

SCATTERING OF AN EXPONENTIAL PULSE BY A SINGLE ATOM

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Abstract. We discuss the scattering of a light pulse by a single atom in free space using a purely semi-classical framework. The atom is treated as a linear elastic scatterer allowing to treat each spectral component of the incident pulse separately. For an increasing exponential pulse with a dipole radiation pattern incident from full solid angle the spectrum resulting from interference of incident and scattered components is a decreasing exponential pulse.*

1. INTRODUCTION

Recent experiments devoted to the coupling of single two-level emitters to the light field in free space have been mainly concerned with the scattering of monochromatic coherent laser beams [1–6]. In these experiments the incident light field has been so weak that the steady-state population of the emitter's excited state was negligible.

Unlike in the above cases, experiments with non-monochromatic incident light fields have been performed in Refs. [7,8]. In Ref. [7] the incident light field was constituted by a constant stream of single photons from a source molecule, *i.e.* pulses with an exponentially decaying temporal envelope having a spectral width matching the one of the transition of the target molecule. These pulses are expected to create a non-negligible excited state population [9]. However, this quantity has not been measured in Ref. [7]. Instead, the extinction of the photon stream has been monitored. In Ref. [8] the incident radiation was a coherent state light pulse with an increasing exponential envelope and a finite amount of atomic excitation was measured.

Here, we want to establish a link between elastic scattering experiments – usually prohibitive of atomic excitation – and the absorption of single photons or weak

*This paper is dedicated to Professor Valentin Vlad at the occasion of his anniversary in 2013. His merits in promoting science in Romania are widely recognized and appreciated.

coherent state pulses. To do so, we treat the atom as a driven harmonic oscillator with a driving force that is weak enough to keep the oscillator's response in the linear regime. This approach is motivated by the close analogy between a coherently driven classical harmonic oscillator and a single atom driven by a single photon [10]. We will decompose a light pulse into its spectral components. Each of these components constitutes a monochromatic wave. It is assumed that the scattering of each of these waves is completely elastic, enabling interference with the corresponding incident spectral component. The resulting spectrum then determines the temporal response of the atom.

Of course, this treatment is not applicable to cases where more than a single photon is contained in the incident pulse. As is evident from fully quantum mechanical treatments [11], the upper state population and hence the electromagnetic field will exhibit Rabi oscillations. This is clearly not covered by the treatment discussed in this paper. However, even a pulse containing the energy of a single photon resonant with the atomic transition induces a non-negligible amount of excited state population. For such a pulse with an effective length of the excited state lifetime, the Rabi frequency can be as large as twice the spontaneous emission rate [12]. This corresponds to a saturation parameter of $S = 8$, *i.e.* an excited state population of $\rho_{ee} = 4/9$. This finding suggests that the calculations presented below are only meaningful for pulses containing much less than a single photon, *e.g.* strongly attenuated coherent states as prepared in Refs. [8, 13].

However, the value of ρ_{ee} obtained from S is a steady state quantity. On the time scale of the excited state lifetime – and single photon pulses used in free space experiments are typically of this duration – the steady state is not yet reached. Rather the excited state population has yet to build up from zero. Therefore, one could expect that the fully elastic treatment is justified during almost the complete duration of the pulse, especially if the amplitude of the incident pulse itself increases slowly. This is the case for exponentially increasing pulses, which have been predicted to excite an atom with full efficiency [9, 11, 14].

The paper is organized as follows: In the next section we will briefly revisit the scattering of a monochromatic wave. Then in Sec. 3 this framework will be applied to all spectral components of an exponentially increasing pulse, yielding the spectrum of the temporal atomic response. The paper will close with a brief discussion.

2. SCATTERING BY AN ATOMIC DIPOLE

At first, the scattering of a monochromatic wave by an atomic dipole is reviewed. The atom is taken as a two-level system and considered to be in the steady state under the monochromatic driving field. The derivation of the respective formu-

las is given in Ref. [15]. Here we just recall the results relevant to this paper.

The power scattered by the atom is given by

$$P_{\text{sc}} = \frac{4P \cdot \Omega \eta^2}{(4\Delta^2/\Gamma^2 + 1)(1 + s)^2} \quad (1)$$

P is the power of the incident beam. Ω is the solid angle of the focused field weighted by the angular intensity pattern $I(\vartheta, \phi)$ of the atomic transition dipole moment [16]:

$$\Omega = \frac{\int_{\phi_{\text{foc}}} \int_{\vartheta_{\text{foc}}} I(\vartheta, \phi) \sin \vartheta d\vartheta d\phi}{8\pi/3} \quad (2)$$

It is given as a normalized quantity ($0 \leq \Omega \leq 1$) with the case $\Omega = 1$ corresponding to focusing from full solid angle. η is the spatial mode overlap of the incident field with the field emitted by the atomic dipole. The overlap is integrated over and normalized to only the part of the solid angle covered by the incident beam. $\Delta = \omega - \omega_0$ is the detuning from the atomic resonance ω_0 , and Γ is the spontaneous emission rate. Finally, s is the saturation parameter which depends on all of the other parameters given above, see Ref. [15]. However, since here we are interested in the regime of elastic scattering we set $s = 0$ and have

$$P_{\text{sc}} = \frac{4P \cdot \Omega \eta^2}{4\Delta^2/\Gamma^2 + 1} \quad (3)$$

This equation is equivalent to the findings of Refs. [17–19], once one identifies the quantity $4\Omega\eta^2$ with the scattering ratio used in these papers.

P_{sc} can be written as $P_{\text{sc}} = \text{const.} \times |E_{\text{sc}}|^2$, where $E_{\text{sc}} = A_{\text{sc}} \cdot e^{i\varphi_{\text{sc}}}$ is the complex amplitude of the dipole wave scattered by the atom. Neglecting proportionality constants, we can write $A_{\text{sc}} = \sqrt{P_{\text{sc}}}$, *i.e.*

$$A_{\text{sc}} = \frac{2A \cdot \sqrt{\Omega} \eta}{\sqrt{4\Delta^2/\Gamma^2 + 1}} \quad (4)$$

where $A = \sqrt{P}$ is the modulus of the incident field amplitude. φ_{sc} is the phase of the the scattered wave *relative* to the phase of the incident field. It is given by [15, 17]

$$\varphi_{\text{sc}} = \arctan\left(\frac{2\Delta}{\Gamma}\right) + \frac{\pi}{2}, \quad (5)$$

as has been confirmed in a recent experiment [20]. With A_{sc} and φ_{sc} we have all quantities at hand that are needed to calculate the field resulting from the superposition of incident field and scattered field.

In almost all of the recent experiments dealing with light-matter interaction in free space, the light scattered by the atom and the incident radiation are collected by optics spanning nominally the same solid angle fraction as the device focusing the incident radiation. The part of the solid angle not covered by the collection optics is

governed by the scattered field alone. The power emitted into this part of the solid angle is

$$P_{\text{sc},\bar{\Omega}} = (1 - \Omega) \cdot \frac{\Gamma^2 \cdot \Omega \eta^2}{\Delta^2 + \Gamma^2/4} \cdot A^2 \quad (6)$$

with the corresponding complex field amplitude

$$E_{\text{sc},\bar{\Omega}} = \frac{\Gamma \eta \sqrt{\Omega(1 - \Omega)}}{\sqrt{\Delta^2 + \Gamma^2/4}} \cdot A e^{i\varphi_{\text{sc}}} \quad (7)$$

The fraction of the scattered power emitted towards the collection optics is

$$P_{\text{sc},\Omega} = \frac{\Gamma^2 \Omega^2 \eta^2}{\Delta^2 + \Gamma^2/4} \cdot A^2 \quad (8)$$

In this solid angle fraction, where the scattered light interferes with the rediverging incident light, an additional $\pi/2$ shift related to the Gouy phase has to be considered [4, 6, 17, 21]. We do this by writing

$$\varphi_{\text{sc}} = \arctan\left(\frac{2\Delta}{\Gamma}\right) + \pi \quad (9)$$

The corresponding field amplitude is

$$E_{\text{sc},\Omega} = \frac{\Gamma \eta \Omega}{\sqrt{\Delta^2 + \Gamma^2/4}} \cdot A e^{i\varphi_{\text{sc}}} \quad (10)$$

We assume that all of the incident radiation is collected as well. However, only a part of the scattered radiation can interfere with the incident field. The corresponding power fraction is proportional to η^2 . Thus, the field component due to interference is

$$E_{\Omega,\text{coh}} = \frac{\Gamma \eta^2 \Omega}{\sqrt{\Delta^2 + \Gamma^2/4}} \cdot A e^{i(\varphi_{\text{sc}} + \varphi_0)} + A e^{i\varphi_0} \quad (11)$$

where we have also allowed for some arbitrary relative phase φ_0 of the incident field. The corresponding power reads

$$P_{\Omega,\text{coh}} = \left[1 + \frac{\Gamma^2}{\Delta^2 + \Gamma^2/4} (\Omega^2 \eta^4 - \Omega \eta^2) \right] \cdot A^2 \quad (12)$$

For completeness, we also give the respective expressions for the fraction that does not interfere with the incident field. The complex field amplitude reads

$$E_{\Omega,\text{incoh}} = \frac{\Gamma \Omega \eta \sqrt{1 - \eta^2}}{\sqrt{\Delta^2 + \Gamma^2/4}} \cdot A e^{i\varphi_{\text{sc}}} \quad (13)$$

with the corresponding power

$$P_{\Omega,\text{incoh}} = \frac{\Gamma^2 \Omega^2 \eta^2 (1 - \eta^2)}{\Delta^2 + \Gamma^2/4} \cdot A^2 \quad (14)$$

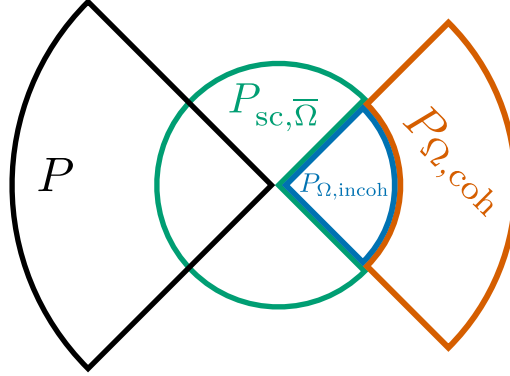


Fig. 1 – Illustration of the different power fractions involved in elastic scattering by a single atom. The incident field is focused from a solid angle fraction Ω with power P . In transmission, the incident field interferes with the scattered field, with the solid angle fraction on which the interference occurs again being Ω . Since the spatial mode overlap between scattered and incident field is in general not perfect, one has to account for a power fraction $P_{\Omega,\text{coh}}$ due to this interference and a remaining fraction of the scattered light with power $P_{\Omega,\text{incoh}}$. The light scattered into part of the solid angle complementary to the transmission one is of power $P_{\text{sc},\bar{\Omega}}$.

With some algebra it is easy to check that energy is conserved. The meaning of the different power fractions is illustrated in Fig. 1.

3. INCIDENT PULSE WITH INCREASING EXPONENTIAL ENVELOPE

In the following we treat the case of an incident wave with carrier frequency ω_0 and an exponentially increasing intensity envelope. The time constant of the exponential shall be the life time of the upper atomic state and the wave ends at $t = 0$. The time dependent field amplitude of such a wave is given by $A_0 \cdot \exp(\Gamma t/2) \cdot H(-t)$ with $H(t)$ being the step function. The spectrum of this wave is given by

$$S(\Delta) = A_0 \cdot \frac{1}{\Gamma/2 + i\Delta} \quad (15)$$

with the spectral amplitude $A(\Delta) = A_0/\sqrt{\Delta^2 + \Gamma^2/4}$ and the relative spectral phase $\varphi_0(\Delta) = \arctan(-2\Delta/\Gamma)$.

For this incident pulse spectrum the resulting spectral field amplitudes can be written as

$$E_{\text{sc},\bar{\Omega}}(\Delta) = A_0 \cdot i \frac{\eta\Gamma\sqrt{\Omega(1-\Omega)}}{\Delta^2 + \Gamma^2/4} \quad , \quad (16)$$

$$E_{\Omega,\text{incoh}}(\Delta) = A_0 \cdot i \frac{\Gamma\Omega\eta\sqrt{1-\eta^2}}{\Delta^2 + \Gamma^2/4} \quad , \quad (17)$$

$$E_{\Omega,\text{coh}}(\Delta) = A_0 \cdot \frac{\Gamma(1/2 - \Omega\eta^2) - i\Delta}{\Delta^2 + \Gamma^2/4} . \quad (18)$$

The last term can be rewritten as

$$E_{\Omega,\text{coh}}(\Delta) = A_0 \times \left[(1 - \Omega\eta^2) \cdot \frac{\Gamma/2 - i\Delta}{\Delta^2 + \Gamma^2/4} + \Omega\eta^2 \cdot \frac{-\Gamma/2 - i\Delta}{\Delta^2 + \Gamma^2/4} \right] . \quad (19)$$

The first term of this sum is again the spectrum of the incident increasing exponential pulse but weighted with a proportionality factor $(1 - \Omega\eta^2)$. The second term which scales with $\Omega\eta^2$ is also the spectrum of an exponential pulse, but for one with a decaying envelope that starts at $t = 0$. In other words, the spectral components of the latter term are the phase conjugated versions of the incident components. Although we have assumed that the scattering is completely elastic, *i.e.* there is no upper state population, we interpret this exponentially decaying fraction as absorbed and spontaneously re-emitted photons.

The spectral amplitudes in Eqns. (16) and (17) correspond to a field envelope that is increasing with $\exp(\Gamma/(2t))$ for $t < 0$ and decreasing with $\exp(-\Gamma/(2t))$ for $t > 0$. This is obvious from the expansion $\Gamma = (\Gamma/2 + i\Delta) + (\Gamma/2 - i\Delta)$ in the numerator of these equations, yielding the sum of the spectra corresponding to the above temporal pulse shapes. The interpretation of this spectrum is straightforward: As long as the incident pulse is nonzero, $E_{\text{sc},\bar{\Omega}}$ is given by the elastically scattered incident wave with increasing exponential envelope. For $t > 0$ there is no incident field amplitude, hence the exponential decay is again interpreted to 'mimic' spontaneous emission. The same interpretation applies to $E_{\Omega,\text{incoh}}$. In a recent experiment [8] the light scattered by a single atom into the backward solid angle $(1 - \Omega)$ has been measured for focusing from $\Omega = 0.11$. The temporal evolution obtained for exponentially increasing coherent state pulses containing three photons on average resembles the double sided exponential corresponding to the spectrum in Eq. (16).

Summing up the power of all exponentially decaying contributions of $E_{\Omega,\text{coh}}$, $E_{\Omega,\text{incoh}}$ and $E_{\text{sc},\bar{\Omega}}$ and normalizing to the total power shows that the power fraction in the temporally decaying signal is given by $\Omega\eta^2$. This suggests to interpret $\Omega\eta^2$ as the absorption probability of an exponentially increasing pulse that is temporally 'mode-matched' to the atomic transition, which is in accordance with the findings of Ref. [13].

In the limit of focusing from full solid angle, $\Omega = 1$, $E_{\text{sc},\bar{\Omega}}$ naturally vanishes. If then also the spatial mode matching becomes perfect, $\eta = 1$, also $E_{\Omega,\text{incoh}}$ and the first term of $E_{\Omega,\text{coh}}$ become zero. In other words, the remaining spectral field components are completely given by the spectrum of an exponentially decaying field. This is the same result one would obtain from a fully quantized treatment of the absorption of a single photon by a single atom, if the incident single photon is the time reversed version of a spontaneously emitted photon [9]. We note that the results

obtained here are also analogous to the ones obtained for the response of an empty Fabry-Perot resonator [10], which constitutes a fully classical problem.

4. DISCUSSION

By using a fully elastic treatment in describing the scattering of light by single atoms, we have derived the electric field spectrum arising from scattering an increasing exponential pulse. The obtained results suggest to interpret the response as a field arising *via* spontaneous emission, which only occurs after absorption of a photon. Recent experiments [8] confirm the expected validity of our framework in the regime of low average photon numbers. Nevertheless, one has to take into account that in the reported experiment roughly 11% of the solid angle was used for focusing. This means that also the coupling efficiency is limited by this value [12, 13]. Therefore, it is not surprising that qualitative agreement with the model presented here is observed, since approximately only every ninth photon interacts with the atom. It will therefore be interesting to compare our theory to the experiments prepared in Refs. [13, 22], where the coupling will occur from almost full solid angle and with large expected mode overlaps [13]. Using the experimental parameters, the relative magnitudes of the exponentially increasing and decreasing fractions should resemble the achieved absorption efficiency. Moreover, increasing the average photon number of the coherent state pulses used in the experiment will identify the boundary of validity of the fully elastic model presented here.

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