Cooperative emission of a pulse train in an optically thick scattering medium

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An optically thick cold atomic cloud emits a coherent flash of light in the forward direction when the phase of an incident probe field is abruptly changed. Due to cooperativity, the duration of this phenomena can be much shorter than the excited lifetime of a single atom and, surprisingly, it weakly depends on the temperature of the gas and on the probe frequency. Repeating periodically the abrupt change of the incident field phase, we generate a forward transmitted train of pulses with short repetition time. It is even possible to quench single atom fluorescence, transferring almost completely the incident power into the pulse train with a high intensity contrast.

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The seminal work on superradiance of R. Dicke in 1954 has opened up tremendous interest in studying cooperative emission of electromagnetic radiation from an ensemble of radiative dipoles (see \[1\] for original proposal, \[2, 3\] for reviews and \[4–10\] for recent related works). In his original proposal, R. Dicke considered an ensemble of \(N\) excited two-level atoms confined inside a volume smaller than \(\lambda^3\), where \(\lambda\) is the transition wavelength. In this context, a macroscopic polarization is built up in the medium upon incoherent spontaneous emission. This Dicke superradiance mechanism, leads to the coherent emission of an intense pulse with a decay time, \(\tau_D = (N \Gamma)^{-1}\), that is shortened by a factor of \(N^{-1}\) with respect to the atomic excited state lifetime \(\Gamma^{-1}\). For practical implementation in the optical domain, the Dicke model was extended to media with volume larger than \(\lambda^3\) \[2, 11\] \[12\]. In those cases, the propagation of the electromagnetic field in the medium and the spatial mode density must be taken into account. If the medium is dense, i.e. \(\rho \lambda^3 \gg 1\), where \(\rho\) is the radiator spatial density, it still exhibits the main feature of the Dicke superradiance, namely the emission of a short pulse after some delay \(\delta\) \[13, 14\]. It was, however, pointed out in Ref. \[11\], that the superradiant pulse decay time should be corrected as \(\tau = \tau_D / \mu\). \(\mu < 1\) is a geometrical factor corresponding to the solid angle subtended by the superradiant emission \[2\] \[12\].

For a dilute scattering medium, i.e. \(\rho \lambda^3 \ll 1\), the Dicke superradiance mechanism does not occur \[15\]. But, an optically thick medium driven by a coherent incident field shares interesting similarities with Dicke superradiance; here, the cooperativity factor \(N \mu\) is replaced by the optical thickness of the medium \[16, 17\]. Once a driving coherent field is abruptly switched off, like in a free induction decay experiment \[18, 26\], a short coherent cooperative flash of light is emitted in the forward direction. The flash duration is inversely proportional to the optical thickness and the bare linewidth of the transition \[24\].

In a coherently driven medium, the incident probe frequency can be detuned with respect to the atomic resonance, leading to non trivial phase rotation of the cooperatively emitted field \[20\]. Moreover, the flash effect can also occur by abruptly changing the phase of the probe field, leading to an interference between the flash and the probe field \[19, 27\]. In this Letter, we largely explore these peculiarities to generate high repetition rate pulse trains in an optically thick cold dilute atomic ensemble, using the setup schematically shown in Fig. 1a. An example of a pulse train, generated in our experiment by periodically changing the probe phase, is shown in Fig. 1b. As a direct proof of cooperative emission, we note that the repetition time, \(T_R\), of the pulse train is clearly shorter than the atomic excited state lifetime \(\Gamma^{-1}\). Moreover, we show that at high repetition rate, the single atom fluorescence is quenched. This constitutes a rather counterintuitive result, with practical importance, addressed in this Letter.

The scattering medium is a cloud of laser-cooled \(^{88}\text{Sr}\) atoms (see Refs. \[20, 28\] for details of the cold atoms production line). The ellipsoidal shape of the cold cloud has an axial radius of 240(10) \(\mu\)m and an equatorial radius of 118(5) \(\mu\)m, with peak density around \(4.6 \times 10^{11}\) cm\(^{-3}\) – thus \(\rho \lambda^3 \approx 0.15\), in the dilute regime – for a total of \(2.5(5) \times 10^8\) atoms. \(\lambda = 689\) nm is the wavelength associated to the \(^{1}\text{S}_0 \rightarrow {^3}\text{P}_1\) intercombination line (bare linewidth of \(\Gamma / 2\pi = 7.5\) kHz) used in this experiment. The temperature of the cold gas is \(T = 3.3(2)\) \(\mu\)K. We get \(k \tilde{v} = 3.4\Gamma\), signaling a significant Doppler broadening of the narrow intercombination line. \(k = 2\pi / \lambda\) is the wavevector of the transition, and \(\tilde{v} = \sqrt{k_B T / m}\) is
In the frequency domain, is given by:

\[ E_t(\omega) = E_0(\omega) \exp \left[ i \frac{n(\omega) \omega L}{c} \right]. \tag{1} \]

In the above equation, \( n(\omega) \), \( E_0 \), \( c \) and \( L \) are the complex effective refractive index, the incident optical field, the speed of light in vacuum and the slab thickness along the laser beam respectively. For a dilute medium, \( n(\omega) = 1 + \rho \alpha(\omega)/2 \) [29], with the two-level atomic polarizability,

\[ \alpha(\omega) = -\frac{3\pi \Gamma c^3}{\omega^3} \int_{-\infty}^{+\infty} dv \exp \left( -v^2/2\bar{v}^2 \right) \delta - kv + i\Gamma/2. \tag{2} \]

\( \delta = \omega - \omega_0 \) is the detuning of the probe laser frequency \( \omega \) with respect to the bare atomic resonance frequency \( \omega_0 \). The effect of Doppler broadening is included in the polarizability by averaging over the thermal Gaussian distribution of the atomic velocity \( v \) along the beam propagation direction. Following the calculation method in Ref. [29], and performing an inverse Fourier transform, we can compute the transmitted intensity as a function of time, \( I_t(t) \). At this stage, we define the optical thickness \( b_\delta(\delta) \) and the relative phase between the transmitted and the incident fields \( \theta_\delta(\delta) \) at the probe detuning and for a given \( \bar{v} \) by

\[ b_\delta(\delta) = \frac{2\omega}{c} \Im [n(\omega)] L, \quad \theta_\delta(\delta) = \frac{\omega}{c} \Re [n(\omega)] - 1 |L|. \tag{3} \]

The transmitted field, \( E_t \), results from the interference between the incident field \( E_0 \) and the field scattered in the forward direction \( E_s \),

\[ E_t = E_0 + E_s. \tag{4} \]

Since we work with effective two-level atoms, we drop the vectorial nature of the electric fields and represent them as scalar quantities. Due to the non-instantaneous response time of the medium, the coherent scattered field in the forward direction is a continuous function across the abrupt change of the incident field. Notably, in a free induction decay experiment where the incident field is abruptly switched off, say at \( t = 0 \), the intensity of the transmitted field at \( t = 0^+ \) is a direct measurement of the forward scattered intensity in the stationary regime. Its properties are studied in detail in Refs. [25, 26]. In particular, following energy conservation arguments, it is shown in Ref. [26] that the intensity of the forward scattering is bounded by 4 times the incident intensity (“superflash effect”). The temporal evolution of the transmitted field, after the abrupt switch off of the incident field, is not a simple function having only one characteristic decay rate [24]. However, we get a clear physical insight considering only the initial lifetime (at time \( t = 0^+ \)), which takes a simple analytical expression (see the rms velocity of the gas. Here, \( k_B \) is the Boltzmann constant and \( m \) is the atomic mass. As an important consequence, the optical thickness strongly depends on temperature. At resonance, we measure \( b_\bar{v}(0) = 19(2) \) along the equatorial axis of the cloud.

A 150 \( \mu \)m diameter probe laser beam, tuned around the intercombination line, is sent through the cold atomic gas along an equatorial axis. The probe power is 400(40) pW, corresponding to 0.45(5)\( I_{\text{sat}} \) (\( I_{\text{sat}} = 3 \) \( \mu \)W/cm\(^2 \)). We measure the forward transmitted intensity of the probe using a photodetector, integrating over the transverse dimensions of the transmitted beam. We apply a bias 1.4 G magnetic field along the beam polarization during the probing phase, making the atom an effective two-level system on the \( ^1S_0 \rightarrow ^3P_1 \) transition.

The ellipsoidal shape of the cloud is modeled by a slab geometry, so that the coherent transmitted electric field,
Supplemental Material [30]:

\[ \tau_\text{b}(\delta) = \left| \frac{I_b(t = 0^+) - I_b(t = \infty)}{dI_b/dt(t = 0^+)} \right| = \frac{2}{\Gamma b(0)} \frac{1 + \exp(-b) - 2\exp(-b/2)\cos(\theta)}{1 - \exp(-b/2)\cos(\theta)} \]  

(5)

where \( b \equiv b(\delta) \) and \( \theta \equiv \theta(\delta) \). In Eq. (5), \( b(0) \) is the optical thickness at resonance and zero velocity. It is linked to \( b(0) \) by \( b(0) = b(0)(g/kv/\Gamma) \) where \( g(x) = \sqrt{\pi/8} \exp(1/8x^2) \text{erf}(1/\sqrt{8x})/x \) [23]. In our experiments, \( g(kv/\Gamma) = g(3.4) \approx 0.16 \), thus for \( b(0) = 19(2) \), we get \( b(0) = 120(10) \). A direct measurement gives a slightly smaller value, \( b(0) = 95(5) \) (see Supplemental Material [30]). The expression of \( \tau_\text{b} \), given by Eq. (5), simplifies to \( \tau_\text{b}(0) = 2[b(0)(\Gamma)]^{-1} \) at resonance (\( b \gg 1 \), \( \theta = 0 \)) and to \( \tau_\text{b}(\pm \infty) = 4[b(0)(\Gamma)]^{-1} \) far-off resonance (\( b = 0 \) and \( \theta = 0 \)). The solid blue curve in Fig. 2 is a plot of \( \tau_\text{b}(\delta) \) for \( kv/\Gamma = 3.4 \). \( \tau_\text{b} \) weakly depends on \( \delta \), but mainly on \( b(0) \) which can be much larger than the optical thickness \( b(0) \) seen by a resonant probe at finite temperature. This strongly reduces the lifetime of the forward scattered field with respect to the atomic lifetime \( \Gamma^{-1} \). Equation (5) has a rather simple physical interpretation: the second term represents the geometrical properties of the propagation inside the medium (change in amplitude and phase shift) while the term \( 2/\Gamma b(0) \) represents the collective behaviour of all excited dipoles. It does not depend on the atomic velocity, but only on the atomic density integrated along the laser direction, because there is no Doppler effect for photons scattered in the forward direction. Similarly, it does not depend on the detuning because all dipoles acquire the same rotating phase and all decay with the same rate \( \Gamma \) independently of the detuning.

The free induction decay experiment is performed using an acoustooptic modulator (AOM) as a light switching device [see Fig. 1(a)]. The experimental data points, represented by blue open circle in Fig. 2, are in reasonable agreement with the theoretical prediction. The large statistical error bars are mainly due to the evaluation of the \( dI_b/dt(t = 0^+) \) values. The slight positive systematic error, also associated to the determination of \( dI_b/dt(t = 0^+) \), comes from the finite response time of our experimental scheme, of the order of 40 ns (≈ 500Γ). To check the latter statement, we use Eqs. (1) and (2) to numerically compute \( I_b(t) \), using the measured incident intensity during probe extinction to determine \( E_0(\omega) \). We then apply, on the numerical signal, the same procedure used to extract \( \tau_\text{b} \) from the experimental data. The result of this procedure, corresponding to the blue dotted line in Fig. 2, perfectly agrees with the experimental data.

Instead of a free induction decay experiment as discussed above, we now consider an abrupt change of the phase of the incident field by \( \pi \) [see Fig. 1(c)], at constant incident intensity. The initial decay time \( \tau_\text{b} \) becomes (see Supplemental Material for the derivation [30]):

\[ \tau_\text{b}(\delta) = \frac{4}{\Gamma b(0)} \frac{1 - \exp(-b/2)\cos(\theta)}{2 - \exp(-b/2)\cos(\theta)} \]  

(6)

We plot this expression as the red dashed line in Fig. 2. If the \( \pi \) phase jump occurs at \( t = 0 \), according to Eq. (4), we have \( E_0(t = 0^+) = -E_0(t = 0^-) + E_s(t = 0^-) \). We aim to observe the largest possible amplitude of the transient field, thus we choose the probe frequency detuning such that the interference between \( E_0(t = 0^-) \) and \( E_s(t = 0^-) \) is destructive. This condition is essentially fulfilled when the incident field is at resonance. If \( b(0) \gg 1 \), \( |E_s(t = 0^-)| \approx |E_0| \), so we expect a coherent flash with a peak intensity \( I_f = 4I_0 \). The destructive interference condition may also happen at a nonzero detuning if the phase rotation experienced by the forward scattered field, \( E_s \), is large enough, for example if \( b(0) \gg 1 \). Under our experimental conditions, this situation occurs at \( |\delta| = 11.3\Gamma \). This corresponds to the superflash regime discussed in [26]. In this context, \( |E_s(t = 0^-)| \approx 1.8|E_0| \), thus the flash has a peak intensity \( I_f \approx (1 + 1.8)^2I_0 \approx 7.8I_0 \). This value is slightly below the maximum value of 9I_0 allowed by energy conservation. The latter value is achievable at larger optical thickness.

The phase change operation is performed using an electrooptic modulator (EOM) placed on the probe laser...
path [see Fig. 1(a)]. The EOM is driven with a high voltage controller having a slew rate of \( \approx 2.3 \text{rad } \mu \text{s}^{-1} \). The two experimental values (red squares), corresponding to \( \delta = 0 \) and \( \delta = 11.3 \Gamma \), are shown in Fig. 2. They are systematically higher than the theoretical prediction for an abrupt phase shift change because of the slow response time of the EOM driver. Similar to free induction decay experiment, we use the experimentally measured EOM driver output to numerically compute the \( I_i(t) \) signal. The resulting values of the decay time (red dash-dotted line in Fig. 2) agree with the experimental ones.

Finally, the last configuration analysed in this Letter consists in applying a square periodic \( \pi \) phase jump. We observe a pulse train with a repetition time \( T_R \) [see an example in Fig. 1(b)] limited by the relaxation time of the system. The cooperative emission in the forward direction dramatically decreases the repetition time below the atomic excited state lifetime.

Bringing the probe on resonance, we plot in Fig. 3(a) (red dots and solid curve) the intensity contrast \( I_c \) of the pulse train. We define \( I_c = \max \{ I_i \} - \langle I_i \rangle \) as the difference between the maximum intensity, \( \max \{ I_i \} \), and the mean intensity, \( \langle I_i \rangle = \frac{1}{T_R} \int_{T_R} I_i(t) \, dt \). We observe an excellent agreement between the experiment and the theoretical prediction of Eqs. (1) and (2). At long repetition time, i.e. \( T_R \gg \Gamma^{-1} \), the system reaches its steady state before the phase jump. Hence, we measure \( I_c \simeq 4 I_0 - \langle I_i \rangle \approx 4 I_0 \). We note that \( \langle I_i \rangle \approx 0 \) [see blue open circles and dashed curve in Fig. 3(a)], most of the incident power is scattered out by single atom fluorescence events. In the \( \tau_0 \lesssim T_R \lesssim \Gamma^{-1} \) intermediate regime, \( I_c \) oscillates and can reach a larger value. Moreover, the mean intensity \( \langle I_i \rangle \) rapidly increases to its maximal value, \( I_0 \). Here, the incident power is almost perfectly transferred to the pulse train. This interesting result can be understood considering cooperativity in forward scattering. Indeed, its characteristic relaxation time scales like \( \frac{b_0(0) \Gamma}{|\delta|} \). Therefore, for \( b_0(0) \gg 1 \), coherent processes relax much faster than single atom fluorescence events. The latter are quenched leading to the good figure of merit at repetition time shorter than \( \Gamma^{-1} \). For \( T_R < \tau_0 \), the repetition rate is faster than any time scale of the atomic ensemble. Even though the probe power is fully transmitted, the contrast \( I_c \) tends to zero.

At detuning \( |\delta| = 11.3 \Gamma \), for long repetition time (\( T_R \gg \Gamma^{-1} \)), the pulses have a higher contrast, \( I_c \simeq (1 + 1.8^2) I_0 - \langle I_i \rangle \approx 7.1 I_0 \) [see Fig. 3(b)]. We note that the large value of the mean intensity observed at long repetition time, namely \( \langle I_i \rangle \approx 0.7 I_0 \), is due to the small optical thickness, \( b_0(\delta) = 0.4 \), at \( |\delta| = 11.3 \Gamma \). Hence, most of the transmitted power is in a continuous transmission mode and not in the pulse train. At intermediate repetition time (\( \tau_0 \lesssim T_R \lesssim \Gamma^{-1} \)), we observe, as a general trend, a decrease of the pulse contrast. Thus, in this regime the figure of merit is generally not as good as for the resonant case.

To conclude, we generate pulse trains of short repetition time using cooperative forward emission in an optically thick scattering medium. We achieve an almost complete transfer of the incident power into the pulse train, quenching the single atom fluorescence, while keeping a high intensity contrast in the pulse. Because the cooperative emission occurs in the forward direction, its decay time weakly depends on the temperature of the gas and on the probe detuning. We employ the narrow intercombination line of strontium as a proof-of-principle, where the time scales are in the order of microseconds. For future practical applications, shorter repetition times in picosecond or sub-picosecond regime should be attainable. For this purpose, one has to use scattering media with higher optical thickness and/or shorter transition lifetime, e.g. thermal vapor of rubidium (\( b_0(0) \approx 600 \) at \( 110 \degree C \)) or a samarium doped fiber (\( b(0) \approx 100 \mu \text{m} \)). The latter allows us to bring this technic in the \( 1.55 \mu \text{m} \) telecommunication band.

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