Creating single collective atomic excitations via spontaneous Raman emission in inhomogeneously broadened systems: Beyond the adiabatic approximation

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The creation of single collective excitations in atomic ensembles via spontaneous Raman emission plays a key role in several quantum communication protocols, starting with the seminal Duan-Lukin-Cirac-Zoller protocol [L.-M. Duan, M. D. Lukin, J. I. Cirac, and P. Zoller, Nature (London) 414, 413 (2001)]. This process is usually analyzed theoretically under the assumptions that the write laser pulse inducing the Raman transition is far off resonance and that the atomic ensemble is only homogeneously broadened. Here we study the impact of near-resonance excitation for inhomogeneously broadened ensembles on the collective character of the created atomic excitation. Our results are particularly relevant for experiments with hot atomic gases and for potential future solid-state implementations.

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I. INTRODUCTION

The creation of collective atomic excitations via spontaneous Raman emission plays an important role in well-known quantum information protocols, in particular the quantum repeater [1] protocol proposed by Duan, Lukin, Cirac, and Zoller (DLCZ) [2]; for other protocols based on the same process see [3–6]. The basic (idealized) scheme is as follows (cf. Fig. 1). In an ensemble of three-level systems with two ground states \( g \) and \( s \) and an excited state \( e \) all \( N \) atoms are initially in the state \( g \). An off-resonant laser pulse on the \( g \)-\( e \) transition (the write pulse) leads to the spontaneous emission of a Raman photon on the \( e \)-\( s \) transition (the Stokes photon). Detection of this photon in the far field, such that no information is revealed about which atom it came from, creates an atomic state that is a coherent superposition of all the possible terms with \( N-1 \) atoms in \( g \) and one atom in \( s \), in the simplest case the completely symmetric state

\[
\frac{1}{\sqrt{N}}\left(|g\rangle|g\rangle\cdots|g\rangle + |g\rangle|s\rangle|g\rangle\cdots|g\rangle + \cdots + |g\rangle|g\rangle\cdots|s\rangle\right).
\] (1)

Such a state corresponds to a single collective atomic excitation in \( s \). In general the term with the \( n \)th atom in \( s \) will have a phase \( e^{i(k_n-k_0)x_n} \), where \( k_n \) is the \( k \) vector of the write laser, \( k_0 \) is the \( k \) vector of the detected Stokes photon, and \( x_n \) is the position of the \( n \)th atom. The phases in Eq. (1) thus correspond to the case \( k_n=k_0 \). Moreover in practice the amplitudes of the different terms may vary, depending on the laser profile and the shape of the atomic ensemble.

A remarkable feature of such collective excitations is that of great interest for practical applications is that they can be read out very efficiently by converting them into single photons that propagate in a well-defined direction, thanks to collective interference [2,7,8]. Resonant laser excitation of such a state on the \( s \)-\( e \) transition (the read laser pulse) leads to an analogous state with \( N-1 \) atoms in \( g \) and one delocalized excitation in \( e \). All the terms in this state can decay to the initial state \( |g\rangle^\otimes N \) while emitting a photon on the \( e \)-\( g \) transition (the anti-Stokes photon). If the phase matching condition \( k_g+k_A=k_g+k_s \) is fulfilled, where \( k_g \) is the \( k \) vector of the read laser and \( k_A \) is that of the anti-Stokes photon, then the amplitudes corresponding to the various terms interfere constructively [provided that there are no other effects disturbing the interference, such as atomic motion or the effects studied in this paper (cf. below)], leading to a very large probability amplitude for emission of the anti-Stokes photon in the direction given by \( k_g+k_s \). For atomic ensembles that contain sufficiently many atoms, emission in this one direction can completely dominate all other directions. This allows a very efficient collection of the anti-Stokes photon [7,8].

Note that there is no such collective interference effect for the emission of the Stokes photon since its emission by different atoms corresponds to orthogonal final states, e.g., the state \( |s\rangle|g\rangle\cdots|g\rangle \) if the Stokes photon was emitted by the first atom, etc. Full “which-way” information about the origin of the photon is thus stored in the atomic ensemble, making interference impossible [9]. As a consequence the total emission probability for the Stokes photon is simply given by the

![FIG. 1. Basic level scheme for the creation of collective atomic excitations in atomic ensembles via spontaneous Raman emission. All atoms start out in \( g \). A laser pulse off-resonantly excites the \( g \)-\( e \) transition, making it possible for a photon to be emitted on the \( e \)-\( s \) transition (with small probability).](image-url)
sum of the emission probabilities for each atom, and there is no preferred direction of emission.

The creation of collective excitations via spontaneous Raman emission is usually analyzed theoretically under the assumption that the write pulse is far off resonance [2,10] (but see Sec. VIB in Ref. [11]). Under this condition it is possible to adiabatically eliminate the excited state. However, in experiments the far off-resonance condition is frequently not fulfilled. From an experimental point of view it can be advantageous to approach resonance in order to increase the rate for the spontaneous Raman process or in order to avoid exciting nearby levels. For excitation relatively close to resonance it is no longer justified to eliminate the excited state. The precise frequency of the excited state then influences the dynamics [12], and it becomes important to consider the effects of inhomogeneous broadening of the transition between ground and excited states [13], which is significant in many experimental situations, e.g., for hot gases [14–17], where the relevant mechanism is Doppler broadening. (Note that the effects of Doppler broadening are negligible in similar experiments with cold atomic gases [7,8,18].)

Inhomogeneous broadening will also be an essential factor in future experiments with solid-state atomic ensembles, in particular rare-earth-metal-doped crystals [19], where it is due to the crystal environment. These systems are otherwise very attractive candidates for realizing the DLCZ and similar protocols, thanks to their excellent coherence properties. For example, storage times exceeding 1 s have already been demonstrated in such a system for coherent atomic excitations in s created via electromagnetically induced transparency (EIT) [20], and light at the single-photon level has been stored and re-emitted using the “atomic frequency comb” protocol [21,22]. Eisaman et al. [19] proposed to reduce the inhomogeneity in these solid-state systems via spectral tailoring techniques similar to those employed in light storage experiments [20,21,23]. However, such an approach greatly reduces the number of available atoms, making it much harder to write and read the atomic excitations efficiently.

Motivated by these considerations, we here analyze the creation of collective atomic excitations in inhomogeneous systems by spontaneous Raman emission without resorting to the usual adiabatic elimination of the excited state. This makes it possible to quantify the impact of near-resonance excitation in combination with inhomogeneous broadening on the collectivity of the created atomic excitation. We introduce the term collectivity for the fidelity of the created excitation with respect to the ideal state of Eq. (1). This quantifies the degree of collective interference that is possible when reading out a given atomic excitation. It is equal to 1 (corresponding to the possibility of perfect collective interference) for excitations that are created under far off-resonant conditions. It is reduced for near-resonant excitation in inhomogeneous systems. In such systems, atoms closer to resonance with the write laser will have a larger amplitude of emitting a Raman photon. Moreover for near-resonant (nonadiabatic) excitation the excited state plays a role in the Raman process, leading to phases that differ from atom to atom in inhomogeneous systems. These effects perturb the collective interference that is at the heart of the readout process.

The main goal of our present work is the characterization of the collective atomic excitation that is created by the emission and detection of the Stokes photon, with a focus on spectral aspects due to near-resonant write excitation and inhomogeneous broadening. Spatial effects have previously been analyzed in detail in Ref. [10] and propagation effects for the anti-Stokes photon have been analyzed in Ref. [11]. This paper is organized as follows. In Sec. II we study spontaneous Raman emission for a single atom under pulsed excitation. We show that the amplitude for detecting a Stokes photon at a given time is proportional to the amplitude of the atom being in the excited state. In Sec. III we show how the single-atom results can be used to quantify the collectivity for an inhomogeneously broadened atomic ensemble. In Sec. IV we give numerical examples relevant to hot atomic gases and solid-state systems. Section V contains our conclusions and an outlook toward future work.

II. SPONTANEOUS RAMAN EMISSION FOR A SINGLE ATOM UNDER PULSED EXCITATION

Let us first recall the well-known dynamics of a two-level system with levels e and g under pulsed excitation (cf. Fig. 2). Suppose that the system starts out in g and that it is excited by a pulse with Rabi frequency $\Omega(t)$ that is detuned from resonance by $\Delta$. We focus on the case $\int dt \Omega(t) < 1$ (moderate pulse area). Depending on the value of $\Delta$ there are different regimes. For far off-resonance excitation, $\Delta \gg \Omega(t)$, the amplitude to be in g is well approximated by $\Omega(0)/\Delta$, such that the system returns to g after the pulse. This is called the adiabatic regime, where the system always stays in the momentarily lowest-energy eigenstate of the Hamiltonian (cf. the lowest curve in Fig. 2). For smaller values of $\Delta$ the population in e attains a maximum during the pulse and then declines, but without returning exactly to zero (cf. the intermediate curve in Fig. 2). Finally, for resonant excitation the population in e increases monotonously for the whole duration (as long as one can neglect the spontaneous decay of the excited level) (cf. the top curve in Fig. 2).

Now add a third level s. We are interested in spontaneous emission on the e-s transition. This means that we have to
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include a continuum of vacuum modes of the electromagnetic field. We follow the Weisskopf-Wigner treatment of spontaneous emission [24]. The relevant states are now $|g⟩|0⟩$, $|e⟩|0⟩$, where $|0⟩$ is the vacuum state of the field, and the set $|s⟩|1ω⟩$, where $|1ω⟩$ is a continuum of one-photon states labeled by the frequency $ω$. For simplicity, and motivated by typical experiments, we focus on one specific direction of emission, treating the effect of the other modes globally as a contribution to the decay of the excited level $e$; note that $e$ can moreover also decay on the $e$-$g$ transition.

Denoting the amplitudes of the states $|g⟩|0⟩$, $|e⟩|0⟩$, and $|s⟩|1ω⟩$ by $α$, $β$, and $γω$, respectively, and the overall decay rate of $e$ by $Γ$, the dynamical equations are

$$\dot{α} = -iΩβ,$$
$$\dot{β} = -iΩα - iΔβ - Γβ - ig ∑_ω γω,$$
$$γω = -iγβ - iωγω. \tag{4}$$

The coupling constant $g$ depends not only on the dipole moment of the $e$-$s$ transition but also on the quantization volume and on the solid angle of the mode under consideration. Its precise form is not important for our purposes here (cf. Ref. [24]). The solution of Eq. (4) is

$$γω(t) = -ig \int_0^t dt' e^{-iω(t-t')} β(t'). \tag{5}$$

Inserting this into Eq. (3) gives

$$\dot{β} = -iΩα - iΔβ - Γβ - g^2 ∑_ω \int_0^t dt' e^{-iω(t-t')} β(t'). \tag{6}$$

Changing the order of summation and integration in the last term [25] and making the approximation

$$\sum_ω e^{-iω(t-t')} ≈ δ(t-t'), \tag{7}$$

we see that the modes under consideration just make a contribution to the overall decay term $-Γβ$, as was to be expected. The relative size of this contribution depends on the solid angle of the considered mode (and also on the branching ratio between the $e$-$g$ and $e$-$s$ transitions). It is essentially negligible in typical experimental situations where the solid angle of collection is small. More interesting for our purposes is the effect of the detection of a Stokes photon at time $t$. Since the annihilation operator for the mode under consideration satisfies $a = ∑_ω δω$ (in the Schrödinger picture), the amplitude for such a detection is

$$c(t) = ∑_ω γω(t) = -∑_ω ig \int_0^t dt' e^{-iω(t-t')} β(t') \propto β(t), \tag{8}$$

where the equality follows from Eq. (5) and the proportionality from Eq. (7). This shows that the amplitude for detecting a Stokes photon is simply proportional to the amplitude of the atom being in the excited state. Below we will show that, as a consequence, the (numerical) solution of the two-level problem gives us all the information we need in order to study the collective interference. For a single atom, the atomic state condition on detecting a Stokes photon is simply $|s⟩$ in the single-atom case. It is more interesting in the $N$-atom case below.

III. COLLECTIVITY IN INHOMOGENEOUSLY BROADENED SYSTEMS

We now consider the situation where the laser pulse excites an ensemble of $N$ atoms, which do not all have the same resonance frequency. Without the third level $s$ one would have a time-dependent state

$$\prod_{n=1}^N [α_n(t)|g⟩_n + β_n(t)|e⟩_n] = \prod_{n=1}^N |G_n(t)). \tag{9}$$

We are interested in the case where there is a third level, but where the spontaneous emission of a photon on the $e$-$s$ transition into the considered directional mode occurs with only a small probability. We then take into account only terms that correspond to a single emission. Detection of a single Stokes photon at time $t$ creates a conditional state proportional to

$$c_1(t)|s⟩|G_2(t)⟩ \cdots |G_{N-1}(t)⟩|s⟩,$$

where for simplicity we again assume $k_5 = k_6$ (no phase factors). The key point is that the coefficients $c_1(t)$ in Eq. (10) are given by the single-atom calculation described in Sec. II. Equation (8) shows that $c_1(t)$ is, in fact, proportional to $β_1(t)$, which depends on the detuning of the corresponding $n$th atom with respect to the laser. The collective atomic state is thus proportional to

$$β_1(t)|s⟩|G_2(t)⟩ \cdots |G_{N-1}(t)⟩|s⟩.$$

After the detection of the considered Stokes photon the states $|G_n(t))$ will continue to evolve, and further photons will be emitted into other directional modes (let us recall that we are interested in the regime where the probability to emit another Stokes photon into the same mode is small). However, in the readout process the corresponding atomic excitations in $s$ will just lead to the emission of additional anti-Stokes photons in other undetected directions. As long as the total number of excitations created in $s$ is much smaller than the total number of atoms $N$, this has no significant effect on the collective interference in the readout process.

Neglecting the additional excitations in $s$ discussed in the previous paragraph, the starting state for the readout is given by

$$β_1(t_s)|s⟩|G_2(t_{sd})⟩ \cdots |G_{N-1}(t_{sd})⟩|s⟩,$$

where $t_s$ is the time when the Stokes photon was detected, whereas $t_{sd}$ is the memory time, i.e., the time when the excitation is read out, which may be much larger than $t_s$. For long enough $t_{sd}$, depending on the lifetime of $e$, all the states $|G_n⟩$ will essentially be equal to $|g⟩$. The state of the atomic excitation in $s$ is then proportional to

$$β_1(t_s)|s⟩|G_2(t_{sd})⟩ \cdots |G_{N-1}(t_{sd})⟩|s⟩.$$
We define the collectivity $C$, which is a function of the time of emission of the Stokes photon $t_S$, as the fidelity of the (normalized) state of Eq. (13) with respect to the ideal state of Eq. (1).

\[ C(t_S) = \frac{\left| \int d\Delta n(\Delta) \beta(t_S, \Delta) \right|^2}{N \int d\Delta n(\Delta) |\beta(t_S, \Delta)|^2}, \]  

where $t_S$ is the time of emission of the Stokes photon as before. The relevant times are therefore those where Stokes photon emission is likely. As we have seen before, the Stokes emission probability for an atom with detuning $\Delta$ at time $t$ is proportional to the population in the excited state, $|\beta(t, \Delta)|^2$. Let us recall that the total emission probability for the Stokes photon can be obtained by summing this quantity over all atoms (there is no collective interference for the Stokes photon emission). It is thus of interest to consider the average excited state population,

\[ p_e(t) = \frac{1}{N} \int d\Delta n(\Delta) |\beta(t, \Delta)|^2. \]  

The values of $t_S$ that are likely to be observed are those for which $p_e(t_S)$ is significant.

For the inhomogeneous broadening we choose $\sigma = 0.5$ GHz, which is a realistic value both for hot gases [14–17,26] and for rare-earth-metal-doped crystals [27]. We furthermore choose $\Omega(t)$ to have Gaussian temporal shape with a full width at half maximum (FWHM) pulse duration of 0.1 $\mu$s, centered at $\tau = 0.2$ $\mu$s in the figures, and with a maximum Rabi frequency of 1 MHz (corresponding to $2\pi \times 10^9$ rad/s), a choice that is again motivated by existing hot gas experiments and by potential future experiments on rare-earth-metal-doped solids.

Figures 4 and 5 show $p_e(t)$ and $C(t_S)$, respectively, for different values of $\Delta_0$. One sees the transition from the far-detuned (adiabatic) regime, where the population (and thus the probability to emit a Stokes photon) follows the Rabi pulse and where the collectivity is high for emission times during the duration of the pulse, to the resonant regime,
where the population no longer follows the pulse and the collectivity is low. The solid lines in the figures correspond to the case $\Gamma=0$ (negligible atomic decay), which is realistic for rare-earth-metal-doped solids, where typical excited state lifetimes are of order 100 $\mu$s to 10 ms [27]. The dashed lines correspond to $\Gamma=5$ MHz, which is a typical value for hot gases. One sees that the collectivity is very similar in both cases, but it stays high a little longer after the pulse in the case with spontaneous decay. Intuitively, the decay of the collectivity after the pulse is due to the fact that Stokes emission for these times is dominated by nonadiabatic contributions (fluorescence) [12]. The spontaneous decay suppresses these contributions compared to the adiabatic ones (Raman scattering) and thus enhances the collectivity. For very early times ($t_0<0.02$ $\mu$s) there are oscillations in the collectivity which we ascribe to a beating between fluorescence and Raman scattering. However, the Stokes photon emission probability for these times is exceedingly low (cf. Fig. 4).

In the absence of other effects the collectivity of a given atomic excitation in $s$ would remain unchanged once the excitation has been created via the emission of the Stokes photon. In practice it will decay on a time scale given by the spin coherence time characterizing the atomic ensemble, which can be very long for solid-state ensembles [20]. The spin transition $g\rightarrow s$ can also be inhomogeneously broadened in these systems, however the associated dephasing can be compensated using spin echo techniques, which have already allowed the demonstration of coherence times as long as 30 s [28]. In the case of hot gases the storage time is also limited by the motion of the atoms [14–16].

Figure 6 shows the quantity $n(\Delta)/|\beta(t_0, \Delta)|^2$ as a function of $\Delta$ for $t_0=0.2$ $\mu$s, i.e., at the center of the pulse. This permits one to see which frequency classes of atoms contribute significantly to the Stokes emission and thus to the collective atomic excitation in $s$ that is created for the different values of $\Delta_0$. One sees that for small $\Delta_0$ only the atoms that are resonant with the laser contribute significantly. On the other hand for large $\Delta_0$ the broad contribution from the bulk of the atomic distribution becomes much more important than that of the resonant atoms (because there are so few of the latter). This is consistent with the results for the collectivity shown in Fig. 5. Intuitively, the sharp feature on resonance corresponds to fluorescence, whereas the broad off-resonant contribution corresponds to Raman scattering [12].

V. CONCLUSIONS AND OUTLOOK

We showed how one can quantify the collectivity of atomic excitations created by spontaneous Raman emission in inhomogeneous ensembles based essentially on simple calculations for a two-level system in combination with suitable averages over the inhomogeneous atomic spectral distribution. We found that quite moderate detuning (of the order of twice the inhomogeneous broadening) is already enough to be in the regime of high collectivity, where collective interference effects are as strong as in homogeneous systems. This is encouraging for future experiments in solid-state systems. It is worth mentioning that these results are in good correspondence with the conditions used in practice in hot gas experiments [14–16]. Of course the precise shape (not just the width) of the atomic distribution has to be taken into account for any given experiment. Note that we have assumed that the inhomogeneous distribution is static, which is an excellent approximation for solid-state atomic ensembles. In hot gases collisions cause frequency changes, which become particularly important for longer write pulses.

In this paper we have focused on the creation of an atomic excitation in $s$, i.e., the write process of the DLCZ protocol. The readout is a priori more complicated because the anti-Stokes emission exhibits collective interference (and thus seems to be less amenable to a single-atom based treatment) and because the anti-Stokes photon can be reabsorbed by the ensemble, in contrast to the Stokes photon that couples to an essentially unpopulated transition. However it should be possible to extend the present approach to a detailed study of the readout as well. The simplest case is a short (with a duration of hundreds of picoseconds or less for the inhomogeneous broadening in our example) intense $\pi$ read pulse that excites all atoms from $s$ to $e$ simultaneously, as described in Sec. I. Once the excitation has thus been transferred to the excited state $e$, the problem is equivalent to the two-level situation studied, e.g., in Refs. [22,29] and should thus be solvable using the same techniques based on the Maxwell-Bloch equations for inhomogeneous systems. Note that efficient readout of excitations in $e$ is possible in inhomogeneous absorbing systems even in the absence of control beams if appropriate phase matching conditions are fulfilled [22,29]. These calculations are typically done in a one-dimensional approximation, which should well describe situations where write and read pulse, Stokes photon, and anti-Stokes photon all propagate along the same axis (in forward or backward direction). However, it may also be possible to extend the three-dimensional descriptions in Refs. [10,30] to the inhomogeneous case.

In practice, short $\pi$ pulses can be hard to implement due to laser power limitations and due to the risk of inducing...
unwanted transitions to nearby levels, where the relevant level separations range from several MHz to several GHz, depending on the system. It is therefore of great interest to investigate alternative readout schemes using chirped pulses. This requires a detailed study of the impact of such excitation schemes on the phases in the collective excitation. EIT effects during the read pulse may also play a role in such a scenario [11]. EIT in inhomogeneous systems has been studied, e.g., in Refs. [31,32].

Let us finally note that inhomogeneous broadening can also have desirable effects. In the context of quantum memory protocols it should allow the efficient implementation of temporal multiplexing [22]. Such “multimode memories” promise great speedups in the context of quantum repeater protocols [33]. It is a fascinating question whether a similar enhancement is possible for the DLCZ protocol.

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[13] In homogeneous systems the collectivity is not affected by excitation close to resonance. However, the excitation has to be sufficiently weak in order to avoid the emission of multiple Stokes photons into the same spatial mode.
[25] The uniform convergence of the final term in Eq. (6), which is required for the reordering of integration and summation to be allowed, can be shown by explicit calculation. This is done notably in the original paper by V. Weisskopf and E. Wigner, Z. Phys. 63, 54 (1930).