Narrowing Spontaneous Emission without Intensity Reduction

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We show that it is possible at least in principle to have unbounded line narrowing in the spontaneous emission spectrum simultaneously with control of the corresponding intensity. For this purpose we employ a laser driven doublet of excited atomic, molecular, or semiconductor states with almost parallel dipole orientation, which is decaying to a ground state. Incoherent pumping is applied to control the population and thus the spontaneous emission intensity of the dressed state associated with the ultranarrow spectral line.

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Spontaneous emission has turned out to be one of the most severe limiting factors for high precision measurements and many novel discoveries in modern quantum optics. Those include effects dependent on a long lifetime of quantum coherence and interferences as high frequency lasing without inversion [1], quantum computing [2], and some mechanisms of laser cooling [3]. One means of affecting fluorescence sensitively is to modify the environmental modal density by a cavity or a band gap material, with strict limitations, however, on the available range of frequencies [4]. Furthermore, one may couple the decaying state of interest to a long living state by a suitable laser field [5]. Here the limitation is the involved weak decay rate and generally further strongly decaying lines arise such that the total fluorescence rate remains unchanged. When two quantum states are close in frequency such that they couple with the same modes of the vacuum, quantum interference comes into consideration [6]. Then particular lines of the resonance fluorescence spectrum may either disappear [7] or exhibit a spectral line with an ultranarrow width [8]. Related schemes have been implemented experimentally in gases [9] and semiconductors [10]. Other systems with very narrow linewidths were put forward [11], but one problem for applications generally is that the narrow lines are not controllable in intensity, in fact generally becoming extremely small in intensity.

The purpose of this Letter is to propose a scheme that supplies both of the following two virtues: it generates a spectral line with an in principle unlimited linewidth, and at the same time it offers a controllable intensity of this line. Thus the narrow line may be weak as in previous suggestions if desired but also rather intense, which is advantageous, for example, for its application as a high precision frequency standard. The scheme put forward here involves a coherently coupled doublet with closely spaced levels and almost parallel dipoles, related to those suggested earlier [7,8,12] with the essential difference of an incoherent pumping among the coherently driven levels. This allows the selective and essentially complete population of any of the dressed states such that the weakly decaying dressed state has the desired population and thus total spontaneous emission intensity. As a further benefit of the incoherent dressed population transfer, otherwise unavoidable additional lines with large width may be rendered to disappear.

The considered system consists of three excited states $|k\rangle$ of energy $\hbar\omega_k$ for $k \in \{a, b, c\}$ and we are interested in modifying their spontaneous emission to a ground state $|g\rangle$ with energy $\hbar\omega_g$ and original rates γ_k for $k \in \{a, b, c\}$. The position between $|a\rangle$ and the closely spaced doublet ($|b\rangle, |c\rangle$) is arbitrary and internal incoherent population transfer in the form of spontaneous emission and incoherent pumping γ_{kl} with $k, l \in \{a, b, c\}$ is included in our approach (see Fig. 1). We retain all incoherent rates; however, we emphasize that many may be chosen zero for a particular choice of system. The transitions from the doublet states to $|g\rangle$ and $|a\rangle$ are envisaged to be dipole allowed while consequently the transition among them is dipole forbidden. The transition rate between $|g\rangle$ and $|a\rangle$, and also between the closely spaced states, will thus be zero or small. A laser field couples the two transitions $a - b$ and $a - c$ equally, with Rabi frequency Ω and laser frequency $\omega = (\omega_{ab} + \omega_{ac})/2$, where $\omega_{jk} = \omega_j - \omega_k$ for $j, k \in \{a, b, c, g\}$. We assume the dipoles of the two doublet states to the ground state $\vec{\mu}_{bg}$ and $\vec{\mu}_{cg}$ to be equal in magnitude but arbitrary in direction. We define the parameter $p = \vec{\mu}_{bg}\vec{\mu}_{cg}/(|\vec{\mu}_{bg}||\vec{\mu}_{cg}|)$ characterizing their

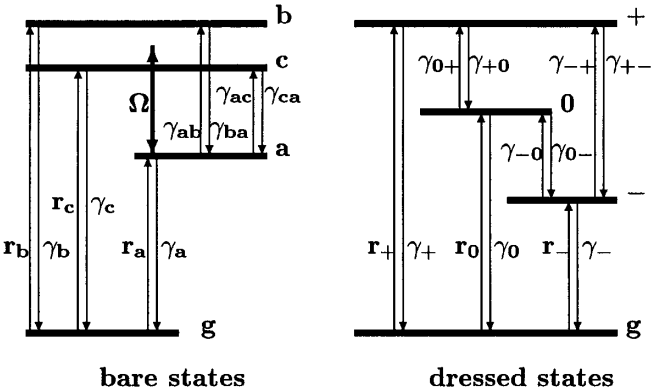

FIG. 1. The bare (left) and dressed state (right) representation of our monochromatically driven four level atom with involved spontaneous decay and incoherent pumping rates.

relative orientation with $p' = \vec{\mu}_{ba}\vec{\mu}_{ca}/(|\vec{\mu}_{ba}||\vec{\mu}_{ca}|)$ being the corresponding parameter for the dipoles to the auxiliary level a. In most cases $p = p'$ and $\vec{\mu}_{ij} = e\langle i|\vec{r}|j\rangle$ for all $i, j \in \{a, b, c, g\}$ with $e\vec{r}$ being the dipole operator. Mostly, we will be interested in situations when p and p' are close to 1 and -1, i.e., the dipoles are almost parallel or almost antiparallel as considered by Zhou and Swain [8]. For later convenience we define $\tilde{p} = 1 - p$ and $\tilde{p}' = 1 - p'$.

We evaluate the spectrum using the method proposed by Cohen-Tannoudji and Reynaud in the dressed states picture [13], which is restricted to strong laser fields, but which allows an analytic and intuitive approach to the problem. For weaker fields the calculation can be easily redone numerically; however, this situation is less favorable because the dressed states are closer to each other with respect to their linewidths and both the trapping of population and the incoherent dressed population transfer are less efficient. Technically, we need to determine the appropriate dressed states, and to calculate the relaxation rates among them and the relaxation rates of the dressed coherences. The coherent part of the Hamiltonian H in the interaction picture reads

$$H = \hbar\delta(|b\rangle\langle b| - |c\rangle\langle c|) + \hbar\Omega(|b\rangle\langle a| + |c\rangle\langle a| + \text{H.a.}) \tag{1}$$

with $\delta = \omega_{bc}/2$. Its eigenstates (dressed states) are

$$\begin{aligned} |0\rangle &= \sqrt{t}\,|a\rangle + \sqrt{s}\,(|c\rangle - |b\rangle), \\ |+\rangle &= \sqrt{s}\,|a\rangle + \tilde{t}|b\rangle + \tilde{s}|c\rangle, \\ |-\rangle &= \sqrt{s}\,|a\rangle - \tilde{s}\,|b\rangle - \tilde{t}|c\rangle, \end{aligned} \tag{2}$$

with $\sqrt{s} = \Omega/G$, $\sqrt{t} = \delta/G$, $\tilde{s} = (G - \delta)/(2G)$, $\tilde{t} = (G + \delta)/(2G)$, $\overline{s} = 2\Omega^4/G^4$, and $G = \sqrt{\delta^2 + 2\Omega^2}$. The polarization of the laser field has been assumed to be half angle between the two dipoles $\vec{\mu}_{ba}$ and $\vec{\mu}_{ca}$ so that the Rabi frequencies on both transitions $a - b$ and $a - c$ can be assumed equal. Including the incoherent relaxation processes as defined in Fig. 1 we obtain the following equations of motion for the density matrix of the system:

$$\begin{aligned}
\dot{\rho}_{aa} &= -(\gamma_a + \gamma_{ab} + \gamma_{ac})\rho_{aa} + \gamma_{ba}\rho_{bb} + \gamma_{ca}\rho_{cc} + r_a\rho_{gg} + p'\sqrt{\gamma_{ba}\gamma_{ca}}\,(\rho_{bc} + \rho_{cb}) \\
&\quad + i[\Omega(\rho_{ab} + \rho_{ac}) - \text{c.c.}], \\
\dot{\rho}_{bb} &= -(\gamma_b + \gamma_{ba})\rho_{bb} + \gamma_{ab}\rho_{aa} + r_b\rho_{gg} - \frac{1}{2}\,(p'\sqrt{\gamma_{ba}\gamma_{ca}} + p\sqrt{\gamma_b\gamma_c}\,)(\rho_{bc} + \rho_{cb}) + i[\Omega\rho_{ba} - \text{c.c.}], \\
\dot{\rho}_{ab} &= -\frac{1}{2}\,(\gamma_a + \gamma_b + \gamma_{ba} + \gamma_{ab} + \gamma_{ac} - i\delta)\rho_{ab} - \frac{1}{2}\,(p'\sqrt{\gamma_{ba}\gamma_{ca}} + p\sqrt{\gamma_b\gamma_c}\,)\rho_{ac} + i\Omega(\rho_{aa} - \rho_{bb} - \rho_{cb}), \\
\dot{\rho}_{bc} &= -\frac{1}{2}\,(\gamma_b + \gamma_c + \gamma_{ca} + \gamma_{ba} + \gamma_{ac} - i\omega_{bc})\rho_{bc} + p'\sqrt{\gamma_{ab}\gamma_{ac}}\,\rho_{aa} + p\sqrt{r_b r_c}\,\rho_{gg} \\
&\quad - \frac{1}{2}\,(p'\sqrt{\gamma_{ba}\gamma_{ca}} + p\sqrt{\gamma_b\gamma_c}\,)(\rho_{bb} + \rho_{cc}) + i\Omega(\rho_{ba} - \rho_{ac}),
\end{aligned} \tag{3}$$

with $\rho_{gg} = 1 - \rho_{aa} - \rho_{bb} - \rho_{cc}$ and corresponding equations for $\dot{\rho}_{cc}$ and $\dot{\rho}_{ac}$ by exchanging b and c in the equations for $\dot{\rho}_{bb}$ and $\dot{\rho}_{ab}$. Explicit time dependences in time T were completely removed by transferring into a rotating frame with $\rho_{ab} = \tilde{\rho}_{ab}\exp[+i\delta T]$ and $\rho_{ac} = \tilde{\rho}_{ac}\exp[-i\delta T]$, and dropping the tilde afterwards.

On using the basis transformation rules [Eqs. (2)], the Hamiltonian in Eq. (1), and the relaxation processes according to Fig. 1 in the bare state basis, we can now evaluate the relaxation processes in the dressed states basis. The dressed states basis is most instructive in the simplifying secular approximation, i.e., when we assume that the Rabi frequency of the driving field Ω is large compared to the largest relaxation rate of the atomic configuration. We then obtain

$$\dot{\rho}_{kk} = -(\gamma_k + \gamma_{kl} + \gamma_{km})\rho_{kk} + r_k\rho_{gg} + \gamma_{lk}\rho_{ll} + \gamma_{mk}\rho_{mm}, \tag{4}$$

with $\{k, l, m\} \in \{+, -, 0\}$ and distinct. The decay rates of the dressed states in the ground state and incoherent pumping rates from the ground states to the dressed states (see right half of Fig. 1 with $\gamma_b = \gamma_c$ and $r_b = r_c$) are given by

$$\begin{aligned} \gamma_0 &= t\gamma_a + 2s\tilde{p}\gamma_b, & r_0 &= tr_a + 2s\tilde{p}r_b, \\ \gamma_+ &= s\gamma_a + \overline{p}\gamma_b, & r_+ &= sr_a + \overline{p}r_b, \end{aligned} \tag{5}$$

with $\overline{p} = 1 - s\tilde{p}$, $\overline{p}' = 1 - s\tilde{p}'$, and $\gamma_+ = \gamma_-$, $r_+ = r_-$. The internal relaxation rates among the dressed states with $\gamma_{ab} = \gamma_{ac}$ and $\gamma_{ba} = \gamma_{ca}$ are

$$\begin{aligned} \gamma_{0+} &= \gamma_{0-} = t\overline{p}'\gamma_{ab} + \overline{s}\tilde{p}'\gamma_{ba}, \\ \gamma_{+0} &= \gamma_{-0} = t\overline{p}'\gamma_{ba} + \overline{s}\tilde{p}'\gamma_{ab}, \\ \gamma_{+-} &= \gamma_{-+} = s\overline{p}'(\gamma_{ab} + \gamma_{ba}). \end{aligned} \tag{6}$$

In the employed strong field regime, the spectrum of resonance fluorescence to the ground consists of three Lorentzian lines with positions $P_k = w_g + \frac{1}{2}(\omega_{bg} + \omega_{cg}) + kG$ for $k \in \{+, -, 0\}$, half widths at half maximum L_k, and weights W_k, arising from the three dressed states $|k\rangle$ for $k \in \{+, -, 0\}$. The positions arise from the magnitudes of the eigenvalues of the Hamiltonian operator to give $+G$, 0, and $-G$. The widths can be found easily as the relaxation rates of the dressed off-diagonal density matrix elements ρ_{kg} for $k \in \{+, -, 0\}$ [13], yielding from the equations of motion

for the population decays with $r = r_a + 2r_b$:

$$L_k = \frac{1}{2}(\gamma_k + \gamma_{kl} + \gamma_{km} + r), \tag{7}$$

with $\{k, l, m\} \in \{+, -, 0\}$ are distinct.

For the evaluation of the weights W_k for the three lines $k \in \{+, -, 0\}$ corresponding to the transitions of the three dressed states to the ground state, we need to solve the population evolution equations (4) in steady state [13], yielding for $\gamma_a = r_b = 0$

$$\begin{aligned} W_0 &= \gamma_0 \rho_{00}(\infty) \\ &= \frac{2s\tilde{p} r_a \gamma_b}{N}[t\overline{p}\gamma_b + \overline{s}\tilde{p}'\gamma_{ab} + t\overline{p}'\gamma_{ba}], \\ W_+ &= W_- = \gamma_+ \rho_{++}(\infty) = \gamma_- \rho_{--}(\infty) \\ &= \frac{r_a \gamma_b \overline{p}}{N}[\overline{s}\tilde{p}\gamma_b + t\tilde{p}'\gamma_{ab} + \overline{s}\tilde{p}'\gamma_{ba}], \end{aligned} \tag{8}$$

with $N = 2s\tilde{p}\overline{p}\gamma_b^2 + \gamma_b[\gamma_{ab}\{2t[1 - s(\tilde{p} + \tilde{p}')] + \overline{s}\tilde{p}'\tilde{p}\} + \gamma_{ba}[2s(t\tilde{p} + 2s\tilde{p}') - \overline{s}\tilde{p}'\tilde{p}] + r_a(t\overline{p} + 2\overline{s}\tilde{p})] + r_a[\gamma_{ab}(2t\overline{p}' + \overline{s}\tilde{p}') + \gamma_{ba}(t\overline{p}' + 2\overline{s}\tilde{p}')]$. Thus we are able to obtain the expression for the full steady state spectrum $S(\omega)$ in the secular approximation in a simple form:

$$S(\omega) = \tau \sum_{k=\{+,0,-\}} W_k \frac{L_k/\pi}{(\omega - P_k)^2 + L_k^2}, \tag{9}$$

with all parameters given in previous equations and τ being a constant depending on the time of measurement.

We begin our discussion with the case when the doublet separation is negligibly small as compared to the Rabi frequency of the driving field such that $t = \delta/G = 0$. The incoherent pumping of the ground state to the excited states r_a enhances the linewidths of interest here and is therefore considered just small enough to supply some excited state population and negligible with respect to all remaining relaxation rates. Assuming also $\tilde{p} = \tilde{p}'$, Eqs. (7) and (8) simplify to

$$\begin{aligned} W_0 &= \frac{\tilde{p} r_a \gamma_{ab}}{(2 - \tilde{p})(\gamma_b + \gamma_{ba}) + \tilde{p}\gamma_{ab}}, \\ W_+ &= W_- = \frac{\frac{1}{2}(2 - \tilde{p}) r_a (\gamma_{ab} + \gamma_b)}{(2 - \tilde{p})(\gamma_b + \gamma_{ba}) + \tilde{p}\gamma_{ab}}, \end{aligned} \tag{10}$$

and

$$\begin{aligned} L_0 &= \frac{1}{2}\tilde{p}(\gamma_b + \gamma_{ba}), \\ L_+ &= \frac{1}{4}\left[(2 - \tilde{p})\left(\gamma_b + \frac{1}{2}\gamma_{ba}\right) + \frac{1}{2}(2 + \tilde{p})\gamma_{ab}\right], \end{aligned} \tag{11}$$

with $L_+ = L_-$. The expression for L_0 in Eq. (11) shows that the linewidth of the central spectral component can be rendered arbitrarily narrow, with the price, however, of a clearly reduced weight [see W_0 in Eq. (10)]. On the other hand, the same equation displays also that this could be corrected by increasing the incoherent process γ_{ab} with respect to γ_b and γ_{ba}. With level a being situated below the doublet, γ_{ab} represents an incoherent pumping rate, i.e., is easily controllable. In Fig. 2 we have plotted the resonance fluorescence spectrum S as a function of the detuning from the central frequency $\frac{1}{2}(\omega_{bg} + \omega_{cg})$. With $\tilde{p}$ approaching 0 we note that the central line becomes significantly narrower. At the same time the weights remained almost constant because the incoherent rate populating the dressed state $|0\rangle$ is increased by a factor of 10, from the dashed to the solid line. For an even larger factor, the total fluorescence could in principle be completely transferred to this central transition ($W_0 \approx r_a, W_+ = W_- \approx 0$). Equally, the sidebands may become extremely narrow and relatively strong for almost antiparallel dipoles and small γ_{ab} with respect to γ_b and γ_{ba}.

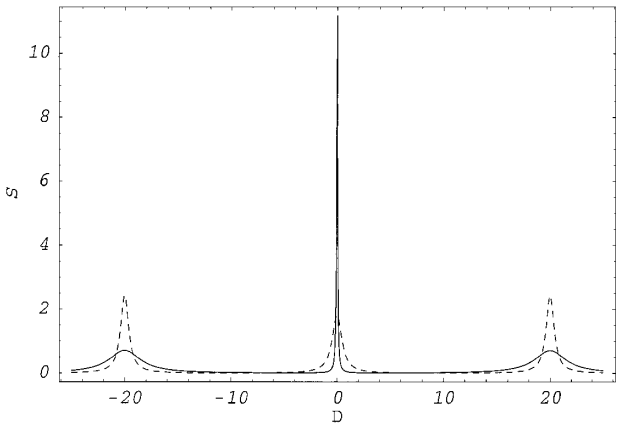

FIG. 2. Spontaneous emission spectrum S to the ground state as a function of the detuning D of the central position $\frac{1}{2}(\omega_{bg} + \omega_{cg})$ from the doublet levels b and c to ground state g for different dipole orientations and incoherent dressed population transfers. The nonvanishing parameters employed are $\Omega = 20/\sqrt{2}$, $\gamma_b = \gamma_c = 1$, $r_a = 10^{-6}$, $\tau = 10^7$; $\gamma_{ba} = \gamma_{ca} = 0.1$ and ω_{bc} negligibly small as compared to G. For the dashed and solid lines the further parameters are $p = p' = 0$; $\gamma_{ab} = \gamma_{ac} = 0.5$ and $p = p' = 0.9$; $\gamma_{ab} = \gamma_{ac} = 5$, respectively.

The spontaneous emission rates among the excited states are given by the particular atomic system chosen and are hard to modify unless one is willing to alter the environmental modal density with a cavity or band gap material. Consequently, in general this spontaneous emission determines a part of the population transfer which in some cases may not be favorable. Therefore the competing incoherent pumping rates among the excited states may remove problems arising from unsuitable spontaneous emission rates. In some cases, however, this is not necessary. In Fig. 3 we consider the case when level a is again below the doublet, where without incoherent pumping $\gamma_{ab} = \gamma_{ac} = 0$. The doublet splitting as opposed to the previous figure is not considered negligible as compared to the driving Rabi frequency. In Fig. 3a we find essentially line narrowing with constant weight $W_0 = r_a t$ and in Fig. 3b even the same for the sidebands with total spontaneous emission cancellation of the central line. Therefore in the latter case the total spontaneous emission

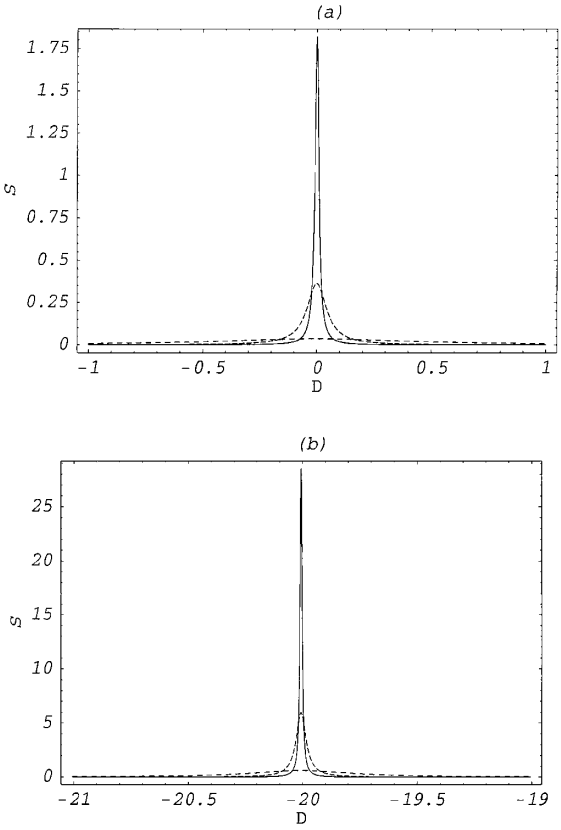

FIG. 3. Spontaneous emission spectrum S of the central (a) and sideband (b) line as a function of detuning D for different dipole orientations. The nonvanishing parameters are $\Omega = 20/\sqrt{2}$, $\gamma_b = \gamma_c = 1$, $r_a = 10^{-6}$, $\tau = 10^6$; $\gamma_{ba} = \gamma_{ca} = 0.1$ and $\omega_{bc} = 2\delta = 1(10)$ for (a) [(b)]. In (a) the central line is plotted for $p = p' = 0$, 0.9, and 0.98 and in (b) one of the two equal outer sidebands for $p = p' = 0$, -0.9, and -0.98 for the dashed (long breaks, almost flat curve), dashed (short breaks), and solid lines, respectively.

may be completely emitted within two arbitrarily sharp lines. The intensity may be further increased with higher pumping rates r_a and r_b, however, then with the consequence of less narrow lines. Also we need to emphasize that the total intensity cannot be enhanced anymore once all of the population is transferred to the dressed state with the small decay rate.

With respect to experimental realization of this effect, related schemes involving quantum interference have recently been implemented [9,10]. The criteria for the choice of system were outlined by Cardimona *et al.* [6], and require especially equal angular quantum numbers and different principal quantum numbers for the doublet states. A possible candidate with proper quantum numbers also for the effects studied here has been realized in the form of a molecule (sodium dimers) [9]. A significant deviation will be the inclusion of incoherent pumping to control the spontaneous emission intensity. This is generally achieved via coherent pumping to an auxiliary state, i.e., with a laser, with successive spontaneous emission to the final state. Here one may prefer a broadband incoherent pump field, colored enough to couple the transition of interest, which was also shown to transfer population and to give rise to quantum interference [14].

In conclusion, we have presented a four-level, coherently driven atomic system where spontaneous emission can be emitted without loss of intensity but with an in-principle unlimited narrow linewidth. The control of the weights of the narrow lines may be established via the choice of a suitable atomic scheme or more flexibly via additional incoherent pumping among the coherently prepared excited states. While the line broadening due to the pumping rate out of the ground state clearly restricts the regime of control of spontaneous emission intensity, the incoherent pumping among the excited states may still be employed to maximize the intensity.

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