Control of spontaneous emission from a coherently driven four-level atom

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We study the spontaneous-emission properties of a coherently driven four-level atom, and show a few interesting phenomena such as fluorescence quenching, spectral-line narrowing, spectral-line enhancement, and spectral-line elimination. These phenomena can be observed in experiment since the rigorous condition of near-degenerate levels with nonorthogonal dipole moments is not required here. Qualitatively, these phenomena can be attributed to the quantum interference between competitive spontaneous-decay channels in the *bare-state picture* or the spontaneously generated coherence between two close-lying levels in the *dressed-state picture*.

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I. INTRODUCTION

Spontaneous emission is well known as a fundamental process resulting from the interaction between radiation and matter. It depends not only on properties of the excited atom but also on the nature of the environment to which the atom is optically coupled. Spontaneous-emission control has attracted much attention because of its potential applications in lasing without inversion $[1]$, transparent high-index materials [2], high-precision spectroscopy and magnetometry $[3]$, quantum information and computing $[4]$, etc. We can achieve the control of spontaneous emission just by putting atoms into different circumstances, such as in free-space, photonic crystals $\lceil 5 \rceil$ and in optical cavities $\lceil 6 \rceil$, which have different densities of electromagnetic modes interacting with atoms. An alternative way to control spontaneous emission is to drive atoms with external coherent fields. Recently, atomic coherence or quantum interference induced by driving fields has become the basic phenomenon for efficient control of spontaneous emission $[7]$. Moreover, it was shown that incoherent decay processes such as atomic spontaneous emission [8] and quantum tunneling of electrons in semiconductor quantum wells [9] also could create atomic coherence or quantum interference, which is usually referred to as spontaneously generated coherence (SGC), decay-induced interference (DII), or vacuum-induced coherence (VIC). Note, however, that SGC only exists in such atoms having two closelying levels subject to the conditions that these levels are near-degenerate and that corresponding dipole matrix elements are not orthogonal. That is, the two close-lying levels should have the same *J* and m_J quantum numbers [10]. It has been predicted that SGC could lead to many interesting phenomena, such as suppression or enhancement of spontaneous emission, selective or total cancellation of fluorescence decay, and narrowing down of spectral linewidth $[11]$. But the rigorous conditions of near-degenerate close-lying levels and nonorthogonal dipole matrix elements are rarely met in real atoms, so few experiments [12] have been carried out to

achieve these interesting phenomena based on SGC. Although it has been pointed out that quantum interference similar to SGC could be achieved in the dressed-state picture of a coherently driven atom without any stringent requirements [13], to our knowledge no theoretical or experimental work has been carried out to study atomic spontaneousdecay properties in the dressed-state picture.

In this paper, we investigate the spontaneous emission spectra of a coherently driven four-level atom both in the bare-state picture and in the dressed-state picture. It is shown that the coherently driven atom, if initially prepared in the coherent population trapping (CPT) state $[14]$, could be trapped in the two ground levels without spontaneously decaying (complete fluorescence quenching) into the metastable level, even if it is interacting with two coherent laser fields. Conversely, if the atom is not in the CPT state at the initial time, there are two spontaneous spectral lines with normal linewidths restricted by spontaneous-decay rates when the CPT condition of two-photon resonance is fulfilled. If the CPT condition of two-photon resonance is slightly deviated, however, we can observe a third, extremely narrow spectral line, which is greatly enhanced or not depending on the initial condition for probability amplitudes of atomic levels. Apart from spectral-line narrowing and enhancement, it is also possible to realize selective fluorescence quenching at one or two special frequency points in the case of a destroyed CPT condition. By qualitative analysis, we show that all the predicted unique phenomena originate from quantum interference between different competitive channels for spontaneous emission in the presence of two coherent driving fields. In the dressed-state picture of one driving field, the interesting phenomena mentioned earlier can also be attributed to SGC. Our proposed scheme for the observation of spontaneous-emission spectra is easy to realize in experiment for most atoms because no specific stringent conditions have to be satisfied.

II. THEORETICAL MODEL AND EQUATIONS

We consider a coherently driven four-level atom in the *Corresponding author: wujinhui0431@sina.com tripod configuration, which consists of one upper, excited

FIG. 1. Schematic diagram of a coherently driven four-level atom, which consists of (a) an upper level and three lower levels in the bare-state picture, and (b) two upper levels and two lower levels in the dressed-state picture of one driving field.

level and three lower, ground or metastable levels [see Fig. 1(a)]. The excited level $|0\rangle$ is respectively coupled to the ground levels $|1\rangle$ and $|2\rangle$ by two coherent laser fields with frequencies (Rabi frequencies) ω_p (Ω_p) and ω_c (Ω_c), while the transition from level $|0\rangle$ to the metastable level $|i\rangle$ is assumed to be coupled by vacuum modes in the free space. Under the rotating-wave and electric-dipole approximations, with the assumption of $\hbar = 1$, the interaction Hamiltonian for the system composed of atom, driving fields, and vacuum modes reads

$$
H_{I} = \sum_{k} \{g_{k0i}e^{j\delta_{k}t}b_{k}|0\rangle\langle i|\} + \Omega_{p}e^{j\Delta_{p}t}|0\rangle\langle 1| + \Omega_{c}e^{j\Delta_{c}t}|0\rangle\langle 2|
$$

+ H.c., (1)

where $b_k^+(b_k)$ is the creation (annihilation) operator for the *k*th vacuum mode with frequency ω_k . $\delta_k = \omega_{0i} - \omega_k$ and g_{k0i} denote the detuning and the coupling constant between the *k*th vacuum mode and the resonance labeled $|0\rangle \leftrightarrow |i\rangle$. Δ_p $=\omega_{01}-\omega_p$ and $\Delta_c=\omega_{02}-\omega_c$ represent detunings of the two coherent driving fields from corresponding resonances.

The state vector for our considered atom at time *t*, whose evolution obeys the well-known Schrödinger equation, can be written as

$$
|\Psi_I(t)\rangle\,{=}\,[a_0(t)|0\rangle\,{+}\,a_1(t)|1\rangle\,{+}\,a_2(t)|2\rangle\,]\big|\{0\}\rangle\,{+}\,\sum_k\,a_{ik}(t)|i\rangle\big|1_k\rangle\,, \eqno(2)
$$

where $|\{0\}\rangle$ denotes the vacuum of the radiation field, and $|1_k\rangle$ means that there is one photon in the *k*th vacuum mode. By using the Weisskopf-Wigner theory $[15]$, we can obtain the following dynamical equations for atomic probability amplitudes $a_i(t)$ in the interaction picture:

$$
\frac{\partial a_0(t)}{\partial t} = -\left(j\Delta_p + \frac{\Gamma_0}{2}\right) a_0(t) - j\Omega_p a_1(t) - j\Omega_c a_2(t),
$$

$$
\frac{\partial a_1(t)}{\partial t} = -j\Omega_p^* a_0(t),
$$

$$
\frac{\partial a_2(t)}{\partial t} = -j(\Delta_p - \Delta_c) a_2(t) - j\Omega_c^* a_0(t),
$$

$$
\frac{\partial a_k(t)}{\partial t} = -j(\Delta_p - \delta_k) a_k(t) - j g_{k0i}^* a_0(t),\tag{3}
$$

where $\Gamma_0 = 2\pi |g_{k0i}|^2 D(\omega_k)$ is the spontaneous-decay rate from level $|0\rangle$ to level $|i\rangle$, and $D(\omega_k)$ is the vacuum-mode density at frequency ω_k in the free space. From Eq. (3), it is easy to find that, only when the two-photon resonance condition $\Delta_p = \Delta_c$ is fulfilled, could we realize coherent population trapping, i.e., have nonvanishing steady-state probability amplitudes $a_1(\infty)$ and $a_2(\infty)$. In the case of $\Delta_p \neq \Delta_c$, it is certain that the atom will finally decay to the metastable level $|i\rangle$.

The spontaneous-emission spectrum is the Fourier transform of $\langle E^-(t+\tau)E^+(t) \rangle_{t\to\infty}$, and can be writen as $S(\delta_k)$ $=\Gamma_0 |a_k(t-\infty)|^2 / 2\pi |g_{k0i}|^2$ for our considered atom. In the following, we use the Laplacian transform method $\lceil 15 \rceil$ and the final-value theorem to obtain

$$
S(\delta_k) = \frac{\Gamma_0}{2\pi} \left[\frac{a_0(0) - \frac{\Omega_p}{\delta_k - \Delta_p} a_1(0) - \frac{\Omega_c}{\delta_k - \Delta_c} a_2(0)}{j\delta_k + \frac{\Gamma_0}{2} + \frac{|\Omega_p|^2}{j(\delta_k - \Delta_p)} + \frac{|\Omega_c|^2}{j(\delta_k - \Delta_c)}} \right]^2.
$$
 (4)

From Eq. (4), we can see that complete quenching of spontaneous emission is possible. The necessary conditions for complete fluorescence quenching are simply $\Delta_p = \Delta_c$, $\Omega_p a_1(0) + \Omega_c a_2(0) = 0$, and $a_0(0) = 0$. In fact, these conditions indicate that the atom should be initially prepared in the CPT state and that the driving fields should be such that the atom cannot be excited to level $|0\rangle$ all the while. The necessary initial atomic state for complete fluorescence quenching can be achieved by the well-developed technique of stimulated Raman adiabatic passage (STIRAP) in experiment [16]. In the general case where $a_0(0) \neq 0$ and $\Delta_p \neq \Delta_c$, however, the spontaneous emission of our considered atom will be quenched at two specific frequencies determined by

$$
\delta_k = \frac{A \pm \sqrt{A^2 - 4Ba_0(0)}}{2a_0(0)},
$$
\n(5)

with $A = [(\Delta_p + \Delta_c)a_0(0) + \Omega_p a_1(0) + \Omega_c a_2(0)$ and *B* $=[\Delta_p \Delta_c a_0(0) + \dot{\Delta}_c \Omega_p a_1(0) + \Delta_p \dot{\Omega}_c a_2(0)].$ Specifically, Eq. (5) will degenerate into $\delta_k = \Delta_p$ and $\delta_k = \Delta_c$ if the atom is initially prepared at level $|0\rangle$ $[a_0(0)=1]$, while if $a_0(0)=0$ and Δ_p $\neq \Delta_c$, it is instead one fluorescence-quenching point that exists in the spontaneous-emission spectra at

$$
\delta_k = \frac{\Delta_c \Omega_p a_1(0) + \Delta_p \Omega_c a_2(0)}{\Omega_p a_1(0) + \Omega_c a_2(0)}.
$$
\n⁽⁶⁾

In the case of $\Delta_p = \Delta_c$, $\Omega_p a_1(0) + \Omega_c a_2(0) \neq 0$, and $a_0(0)$ $= 0$, the numerator of *S*(δ_k) cannot have a value of zero. Thus only when the denominator of $S(\delta_k)$ tends to infinity could we have $S(\delta_k) = 0$ in principle. It is easy to find from Eq. (4), however, that when the denominator of $S(\delta_k)$ tends to infinity at $\delta_k = \Delta_p$, the numerator of $S(\delta_k)$ also tends to infinity, and we have

FIG. 2. Spontaneous-emission spectra $S(\delta_k)$ (in arbitary units) for (a) $\Delta_p = 0.2\gamma$ and $\Delta_c = 0.0$; (b) $\Delta_p = 1.0\gamma$ and $\Delta_c = -1.2\gamma$. Other parameters are $\Omega_c = 0.5 \gamma$, $\Omega_p = 0.3 \gamma$, $\Gamma_0 = \gamma$, $a_0(0) = 1.0$, $a_1(0) = 0.0$, and $a_2(0)=0.0$.

$$
S(\delta_k = \Delta_p) = \frac{\Gamma_0}{2\pi} \left| \frac{\Omega_p a_1(0) + \Omega_c a_2(0)}{\Omega_p^2 + \Omega_c^2} \right|^2.
$$

That is, fluorescence quenching can be cancelled in this special case.

III. RESULTS AND DISCUSSION

In this section, we discuss a few numerical calculations about $S(\delta_k)$ based on Eq. (4). In Fig. 2, we show the general cases where two fluorescence-quenching points exist in the spontaneous-emission spectrum. It is clear that when the two fluorescence-quenching points lie close together, an ultranarrow line can be observed at the center of the spontaneousemission spectrum [see Fig. $2(a)$]. If the two fluorescencequenching points are relatively distant in frequency, two narrow lines instead exist at both ends of the spontaneous emission spectrum [see Fig. 2(b)]. The central ultranarrow line and the narrow sideband lines also could be enhanced, as shown in Fig. 3, under proper initial conditions of probability amplitudes. In Fig. 4, we can find only one fluorescencequenching point in the spontaneous-emission spectrum because we have chosen $a_0(0)=0$ and $\Delta_p \neq \Delta_c$, as discussed in the preceding section. Moreover, there is also an extremely narrow and greatly enhanced fluorescence line near the quenching point. Bear in mind, however, that only if the CPT condition $\Delta_p = \Delta_c$ is slightly deviated, can we observe such an extremely narrow and greatly enhanced spectral line. In the converse case where the CPT condition is badly destroyed, this spectral line (the central one) will be eliminated or become comparable in width to other lines, as shown in

FIG. 3. Spontaneous-emission spectra $S(\delta_k)$ (in arbitary units) for (a) $\Delta_p = 0.2\gamma$ and $\Delta_c = 0.0$; (b) $\Delta_p = 1.0\gamma$ and $\Delta_c = -1.2\gamma$. Other parameters are $\Omega_c = 0.5 \gamma$, $\Omega_p = 0.3 \gamma$, $\Gamma_0 = \gamma$, $a_0(0) = 0.906$, $a_1(0)$ $= 0.3$, and $a_2(0) = 0.3$.

FIG. 4. Spontaneous-emission spectra $S(\delta_k)$ (in arbitary units) for (a) $a_1(0)=1.0$ and $a_2(0)=0.0$; (b) $a_1(0)=0.707$ and $a_2(0)$ = 0.707. Other parameters are $\Omega_c = 0.5 \gamma$, $\Omega_p = 0.3 \gamma$, $\Gamma_0 = \gamma$, Δ_p $= 0.1 \gamma$, $\Delta_c = 0.0$, and $a_0(0) = 0.0$.

Fig. 5. Finally, if $\Delta_p = \Delta_c$, i.e., the CPT condition is fully satisfied, the spontaneous-emission spectra are always doubly peaked with normal width restricted by the spontaneousdecay rate Γ_0 (see Fig. 6).

The interesting phenomena mentioned earlier, including fluorescence quenching, spectral-line narrowing, spectralline enhancement, and spectral-line elimination, can be qualitatively attributed to the quantum interference induced by both driving fields ω_p and ω_c . In the limit of $\Omega_p < \Gamma_0$ and Ω_c < Γ_0 , only one-photon, two-photon, three-photon, and four-photon processes are important and necessary to consider, while other processes involving much more photons can be reasonably ignored. With this assumption, we can see that there exist three groups of competitive pathways for spontaneous emission in our four-level atom, as follows:

(i) The first group of competitive pathways are $|0\rangle \rightarrow |i\rangle$, $|0\rangle \rightarrow |1\rangle \rightarrow |0\rangle \rightarrow |i\rangle$, and $|0\rangle \rightarrow |2\rangle \rightarrow |0\rangle \rightarrow |i\rangle$, corresponding to the four-level atom being initially prepared at level $|0\rangle$. The quantum interference between $|0\rangle \rightarrow |i\rangle$ and $|0\rangle \rightarrow |1\rangle$ \rightarrow $|0\rangle \rightarrow |i\rangle$ will lead to fluorescence quenching at $\delta_k = \Delta_n$, while the quantum interference between $|0\rangle \rightarrow |i\rangle$ and $|0\rangle$ \rightarrow $|2\rangle \rightarrow |0\rangle \rightarrow |i\rangle$ will lead to fluorescence quenching at δ_k $=\Delta_c$. This qualitative conclusion is in agreement with Eq. $(4).$

(ii) The second group of competitive pathways are $|1\rangle$ \rightarrow $|0\rangle \rightarrow |i\rangle$ and $|1\rangle \rightarrow |0\rangle \rightarrow |2\rangle \rightarrow |0\rangle \rightarrow |i\rangle$, corresponding to the atom being initially prepared at level $|1\rangle$. The quantum interference between $|1\rangle \rightarrow |0\rangle \rightarrow |i\rangle$ and $|1\rangle \rightarrow |0\rangle \rightarrow |2\rangle \rightarrow |0\rangle$ \rightarrow *i*) will lead to fluorescence quenching at $\delta_k = \Delta_c$, which is in accordance with Eq. (4).

(iii) The third group of competitive pathways are $|2\rangle$ \rightarrow $|0\rangle \rightarrow |i\rangle$ and $|2\rangle \rightarrow |0\rangle \rightarrow |1\rangle \rightarrow |0\rangle \rightarrow |i\rangle$, corresponding to the atom being initially prepared at level $|2\rangle$. The quantum

FIG. 5. Spontaneous-emission spectra $S(\delta_k)$ (in arbitary units) for (a) $a_0(0)=0.906$, $a_1(0)=0.3$, and $a_2(0)=0.3$; (b) $a_0(0)=0.0$, $a_1(0) = 0.707$, and $a_2(0) = 0.707$. Other parameters are $\Omega_c = 0.5 \gamma$, $\Omega_p = 0.3 \gamma$, $\Gamma_0 = \gamma$, $\Delta_p = 0.4 \gamma$, and $\Delta_c = -0.3 \gamma$.

FIG. 6. Spontaneous-emission spectra $S(\delta_k)$ (in arbitary units) for (a) $\Delta_p = \Delta_c = 0.0$; (b) $\Delta_p = \Delta_c = 0.2 \gamma$. Other parameters are Ω_c = 0.5 γ , Ω_p = 0.3 γ , $\Gamma_0 = \gamma$, $a_0(0)$ = 0.0, $a_1(0)$ = 0.707, and $a_2(0)$ $= 0.707.$

interference between $|2\rangle \rightarrow |0\rangle \rightarrow |i\rangle$ and $|2\rangle \rightarrow |0\rangle \rightarrow |1\rangle \rightarrow |0\rangle$ \rightarrow *i*) will lead to fluorescence quenching at $\delta_k = \Delta_p$, which is also in accordance with Eq. (4).

If we are not sure at which level the atom is initially prepared, all three groups of competitive pathways will simultaneously exist and lead to much more complicated quantum interference so that the fluorescence-quenching points are determined by Eq. (5) or Eq. (6). As for spectralline narrowing, enhancement, and elimination, these phenomena depend on how these competitive pathways interfere and whether the quantum interference is strong. As an example, we consider the special case where the atom is initially prepared at level $|0\rangle$. In this case, the pathway of $|0\rangle$ \rightarrow *i*) involves only a spontaneously emitted photon ω'_k , while the pathway of $|0\rangle \rightarrow |1\rangle \rightarrow |0\rangle \rightarrow |i\rangle$ involves a stimulated-emission photon ω_p , a stimulated-absorption photon ω_p , and a spontaneously emitted photon ω_k . If $\omega_p = \omega_{01}$ $(\Delta_p=0)$, we have $\omega_k = \omega'_k$ and the strongest quantum interference, which then leads to CPT and spectral-line elimination. If ω_p is slightly different from ω_{01} (Δ_p has a small value), ω_k will certainly be slightly different from ω'_k , so the quantum interference becomes a little weak, which then leads to spectral-line narrowing. With the increase of Δ_n , the quantum interference becomes weaker and weaker so that the central narrow spectral line becomes wider and wider. Note that only if the atom is initially prepared at level $|1\rangle$ and/or $|2\rangle$, can we observe spectral-line enhancement in the case of strong quantum interference. That is, spectral-line enhancement depends on both quantum interference induced by driving fields and initial conditions of probability amplitudes.

Next we show an alternative explanation for the preceding interesting phenomena of spontaneous emission. In the dressed-state picture of the driving field ω_c , levels $|0\rangle$ and $|2\rangle$ can be replaced by two new states $|+\rangle$ and $|-\rangle$, whose probability amplitudes are defined as

$$
a_{+}(t) = \cos(\theta)a_{0}(t) + \sin(\theta)a_{2}(t),
$$

$$
a_{-}(t) = \sin(\theta)a_{0}(t) - \cos(\theta)a_{2}(t),
$$
 (7)

with $\tan \theta = \omega_+ / \Omega_c$ and $\omega_{\pm} = (-\Delta_c \pm \sqrt{\Delta_c^2 + 4\Omega_c^2})/2$ representing frequencies of $|+\rangle$ and $|-\rangle$ relative to level $|0\rangle$. Then, Eq. (3) can be rewritten as

$$
\frac{\partial a_1(t)}{\partial t} = -j\Omega_{p+}^* a_+(t) - j\Omega_{p-}^* a_-(t),
$$

$$
\frac{\partial a_+(t)}{\partial t} = -\left(j\Delta_+ + \frac{\Gamma_+}{2}\right) a_+(t) - j\Omega_{p+} a_1(t) - \frac{\Gamma_{+-}}{2} a_-(t),
$$

$$
\frac{\partial a_-(t)}{\partial t} = -\left(j\Delta_- + \frac{\Gamma_-}{2}\right) a_-(t) - j\Omega_{p-} a_1(t) - \frac{\Gamma_{+-}}{2} a_+(t),
$$

$$
\frac{\partial a_k(t)}{\partial t} = -j(\Delta_p - \delta_k) a_k(t) - jg_{k+}^* a_+(t) - jg_{k-}^* a_-(t), \quad (8)
$$

with $\Delta_+ = \Delta_p + \omega_+$, $\Delta_- = \Delta_p + \omega_-$, $\Gamma_+ = \cos^2 \theta \Gamma_0$, $\Gamma_- = \sin^2 \theta \Gamma_0$, Γ_{+-} = sin θ cos $\theta \Gamma_0$, Ω_{p+} = cos $\theta \Omega_p$, Ω_{p-} = sin $\theta \Omega_p$, g_{k+} =cos *gk*0, and *gk*[−] = sin *gk*0. Clearly, there exists quantum interference between the two spontaneous-decay channels $|+\rangle \rightarrow |i\rangle$ and $|-\rangle \rightarrow |i\rangle$ because level $|+\rangle$ interacts with level $|-\rangle$ by spontaneous emission $[(\partial a_+ / \partial t) = \cdots - (\Gamma_{+-}/2)a_-$ and $(\partial a_- / \partial t)$ = ··· - ($\Gamma_{+-}/2$) a_+]. This means that this four-level atom is equivalent to an atom composed of two close-lying upper levels and two well-spaced lower levels [see Fig. $1(b)$] [17], and the quantum interference denoted by Γ_{+-} in Eq. (8) has the same physical meaning as SGC. Thus in the dressedstate picture of field ω_c , we can attribute phenomena such as spectral narrowing, enhancement, and elimination to SGC in the presence of a pumping field ω_p . Because no such stringent requirements as near-degenerate levels and nonorthogonal dipoles have to be fulfilled, corresponding experiments can be easily carried out for real atoms having a level configuration such as that shown in Fig. $1(a)$.

IV. CONCLUSION

In summary, by numerical calculations and qualitative analyses, we have investigated in detail the spontaneousemission spectra of a four-level atom driven by two coherent laser fields in the tripod configuration. It is shown that, by choosing proper parameters, we can observe a few interesting phenomena in the spontaneous-emission spectra, such as total or selective fluorescence quenching, extremely narrow and greatly enhanced spectral lines, and spectral-line elimination. These phenomena critically depend on how the CPT condition of two-photon resonance $(\Delta_p = \Delta_c)$ is fulfilled and the level at which the atom is initially prepared. For example, only if the CPT condition is slightly destroyed and the atom is initially not at level $|0\rangle$ can we observe an extremely narrow and greatly enhanced spectral line in the spontaneous-emission spectra. In the limit of weak driving fields, these interesting phenomena can be seen as resulting from quantum interference between competitive pathways for spontaneous emission. Alternatively, in the dressed-state picture of the driving field ω_c , we also can attribute these interesting phenomena to SGC existing between two closelying and mutually driving levels. It is worth emphasizing that our proposed scheme is suitable for experimental realization because no rigorous atomic conditions, such as neardegenerate levels and nonorthogonal dipoles, have to be satisfied.

Our numerical results for atomic spontaneous emission are most easily experimentally observed in a magneto-optical trap (MOT) [18] where the atomic temperature can be decreased to several tens of μ K so that the Doppler broadening effect can be effectively eliminated. If the atoms are in a cell, however, it is certain that velocity distributions or decay terms due to the interaction with a buffer gas have to be included, and some phenomena described in this paper will be significantly modified, although the underlying physics of quantum interference and atomic coherence is still valid. We

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are currently performing exact calculations on spontaneous emissions from atoms in a cell both with and without a buffer gas.

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