Narrow resonances with excitation of finite bandwidth field

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The effect of the laser linewidth on the resonance fluorescence spectrum of a two-level atom is revisited. The novel spectral features, such as hole-burning and dispersive profiles at line centre of the fluorescence spectrum are predicted when the laser linewidth is much greater than its intensity. The unique features result from quantum interference between different dressed-state transition channels.

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The study of resonance fluorescence spectrum has provided much fundamental insight into the subject of atom-light interactions. It is well known that for weak laser field excitation, the spectrum exhibits a Lorentzian lineshape, whereas it develops into the Mollow triplet for strong field excitation \[1\]. The latter is a direct signature of stimulated emissions and absorptions of the atom during the time interval for one spontaneous decay.

Recently, considerable attention has been paid to modifying the standard resonance fluorescence spectrum. Indeed, there are many ways to achieve this. One method is to place the atom inside a cavity, a wide variety of spectral features, such as dynamical suppression and enhancement, and spectral line narrowing of the Mollow triplet, has been predicted \[2\] and detected \[3\]. Another method is to bathe the atom in a squeezed vacuum. Swain and co-worker \[4\] then predicted anomalous fluorescence spectral features for weak excitation: hole-burning and dispersive profiles at line center, which are \textit{qualitatively} different from any seen previously in resonance fluorescence. Of late, Gawlik \textit{et al.} \[5\] have shown that rapidly elastic collisions between monochromatically driven atoms can also give rise to these anomalous profiles in resonance fluorescence.

In this Letter we report that the anomalous fluorescence spectral features, such as hole-burning and dispersive profiles at line center, can even take place in a system of which a two-level atom is damped by a standard vacuum, and driven by a laser field with a finite bandwidth due to phase diffusions. The effect of the linewidth of the driving laser on the spectrum \[6\] and the intensity fluctuations \[7,8\] of the resonance fluorescence had been extensively investigated both theoretically and experimentally. However, most of these studies concentrated on the case of which the laser intensity is much greater than its linewidth, where the spectral components are well resolved. The spectral line broadening, suppression and asymmetry in the Mollow triplet were reported \[6\]. We are here mainly interested in the region of which the laser linewidth is larger than its intensity. Novel resonance lineshapes, — the hole-burning and dispersive profiles at the spectral line centre of the resonance fluorescence are predicted.

We consider a single two-level atom with transition frequency $\omega_A$ driven by a laser field with amplitude $E$ and frequency $\omega_L$ and fluctuating phase $\phi(t)$. The master equation for the atomic density matrix operator $\rho$, in a frame rotating at the frequency $\omega_L$, is of the form

$$\dot{\rho} = -i [H_{AL}, \rho] + \mathcal{L}\rho,$$

where

$$H_{AL} = \frac{\Delta}{2} \sigma_z + \frac{\Omega}{2} \left[ e^{-i\phi(t)} \sigma_+ + e^{i\phi(t)} \sigma_- \right],$$

$$\mathcal{L}\rho = \gamma (2\sigma_- \rho \sigma_+ - \sigma_+ \sigma_- - \rho \sigma_+ \sigma_-),$$

where $H_{AL}$ is the Hamiltonians of the coherently driven atom and $\mathcal{L}\rho$ describes the atomic spontaneous decay with the rate $\gamma$, $\sigma_\pm$ and $\sigma_z$ are the atomic upper (lower) transition and population inversion operators, respectively,
\( \Omega = 2|\mu_0 E|/\hbar \) is the driving Rabi frequency, \( \Delta = \omega_A - \omega_L \) is the detuning between the atomic transition and the driving laser. The fluctuating phase, \( \phi(t) \), results in a stochastic frequency \( \dot{\phi}(t) = \dot{\phi}(t) \), which is assumed to be a Gaussian random process with the properties \[ E \]  

\[ \langle \dot{\phi}(t) \rangle = 0, \quad \langle \dot{\phi}(t) \dot{\phi}(t') \rangle = 2L\delta(t - t'), \]  

(4) 

where \( L \) is the strength of the frequency fluctuations and physically describes the effective bandwidth of the laser beam due to the phase diffusion. This is the situation most appropriate for describing the radiation from a diode laser, which has a very stable amplitude and very large phase-diffusions when it is operated far above threshold. 

After averaging over the stochastic phase, one can obtain the optical Bloch equation to be \[ E \]  

\[ \langle \dot{\sigma}_- \rangle = -\left( \Gamma + i\Delta \right) \langle \sigma_- \rangle + i\Omega \langle \sigma_z \rangle, \]  

\[ \langle \dot{\sigma}_z \rangle = -\gamma_z \langle \sigma_z \rangle + i\Omega \left( \langle \sigma_- \rangle - \langle \sigma_+ \rangle \right) - \gamma_z, \]  

(5) 

where \( \Gamma = \gamma + L \) and \( \gamma_z = 2\gamma \) represent respectively the transverse and longitudinal relaxation rates. 

The resonance fluorescence spectrum in the far radiation zone can be expressed, in term of the steady-state atomic correlation function, as \[ E \]  

\[ \Lambda(\omega) = \text{Re} \int_{-\infty}^{\infty} \text{lim}_{t \to \infty} \langle \sigma_+(t + \tau) \sigma_-(t) \rangle e^{-i\omega\tau} d\tau = \text{Re}[D(i\omega)], \]  

(6) 

where \( D(z) \) is the Laplace transform of the atomic correlation function \( \lim_{t \to \infty} \langle \sigma_+(t + \tau) \sigma_-(t) \rangle \), which is obtained, by invoking the quantum regression theorem, together with the Bloch equations \[ E \] (when \( L = 0 \)) and Kimble et al. \[ E \] (when \( \Omega \gg L \)). However, here we exploit novel spectral features in the regime of \( L \geq \Omega \), which has been paid little attention in the past. 

Figure 1 presents the resonance fluorescence spectrum of the atom with excitation of a resonant \( \Delta = 0 \), strong laser field \( \Omega = 50\gamma \gg \gamma \) with various laser linewidths \( L = 10\gamma, 50\gamma, 100\gamma, 200\gamma \). It is obvious that when the laser linewidth \( L \) is much less than the Rabi frequency \( \Omega \), see, for example, in the frame \( a \) for \( L = 10\gamma \), as Kimble et al. \[ E \] predicted, the spectrum still exhibits a three-peak structure, but with a suppressed central peak and narrowed sidebands, comparing to the standard Mollow triplet. As the laser linewidth widens, the central peak is greatly suppressed while the sideband resonances are merged, therefore, a dip (i.e., a hole burning profile) places at line centre, —see in Fig. 1(b) where \( L = 50\gamma \). When the value of the laser linewidth is much larger than the Rabi frequency, the dip becomes very narrow, as depicted in Figs. 1(c) and 1(d) where \( L = 100\gamma \) and \( 200\gamma \), respectively. 

Figure 2 illustrates a three dimensional fluorescence spectrum against the laser linewidth, for \( \gamma = 1 \), \( \Omega = 50\gamma \) and \( \Delta = 0 \), from which one can see how the Mollow triplet is suppressed and the dip is developed at line centre as the laser linewidth increases. 

Figure 3 clearly shows that the resonance fluorescence spectrum may exhibit another narrow resonance feature—dispersive (Rayleigh-wing) profile, when the laser with a very wide linewidth is appropriately detuned from the atomic transition frequency. We have taken the parameters \( \gamma = 1 \), \( \Omega = 50\gamma \), \( L = 200\gamma \) in Fig. 3. When the laser-atom detuning is comparable with the laser linewidth, e.g., in Figs. 3(b)-3(c) for \( \Delta = 100\gamma, 200\gamma \), the dispersive profile is most pronounced, otherwise, it is less pronounced, see, for instance, in Fig. 3(a) \( \Delta = 50\gamma \ll L \) and Fig. 3(d) \( \Delta = 400\gamma \gg L \). The latter frame demonstrates the narrow peak at the laser frequency (line centre) and a broad peak at the atomic transition frequency. The both resonances are well split, which is the case of Kimble et al. \[ E \]. 

When the laser bandwidth is much larger than the other parameters, i.e. \( \Gamma \gg \Omega \), \( \Delta \), \( \gamma_z \), the resonance fluorescence spectrum \[ E \] approximately takes the form, 

\[ \Lambda(\omega) \approx \frac{\Gamma}{4(\gamma_z^2 + \Omega^2)} \left[ \frac{\Omega^2 - 2\Delta\omega}{\Gamma^2 + \omega^2} + \frac{\Omega^2 + 2\Delta\omega}{(\Gamma - \Omega^2/\Gamma)^2 + \omega^2} - \left( \frac{\Omega}{\Gamma} \right)^4 \right] \],  

(8) 

which consists of three resonances located at line center, but with different resonance linewidths. The first two resonances have positive weights and linewidths of an order of \( 2\Gamma \) (noting that \( \Omega^2/\Gamma \ll 1 \)), whereas the last one
has a very narrow linewidth of $2\gamma_z$, comparing to $2\Gamma$, and a negative weight. The latter gives rise to a typical spectral profile of which a narrow hole is bored into a broad peak. The approximate expression of the resonance fluorescence spectrum also shows that when the laser is resonant with the atom ($\Delta = 0$), all the three resonances are the Lorentzian lineshape, therefore, the spectrum is symmetry, as shown in Figs. 1-2. Otherwise, these resonances mix up the Lorentzian and Rayleigh-wing (dispersive) lineshapes when the laser is detuned from the atom. As a results, the spectrum exhibits asymmetry, see, for example in Fig. 3.

The hole-burning and dispersive profiles at the spectral centre of the resonance fluorescence are attributed to quantum interference \( \Lambda \). To explain this, we work in the basis of the semiclassical dressed states \(|\pm\rangle\), defined by the eigenvalue equation \( \hat{H}_{AL}|\pm\rangle = \pm(\Omega/2)|\pm\rangle \). For simplicity, we consider only the case of $\Delta = 0$. The dressed states are then given by \(|\pm\rangle = (|0\rangle \pm |1\rangle)/\sqrt{2} \). In the limit of $\Omega \gg \gamma$, the equations of motion then simplify to

\[
\begin{align*}
\langle \dot{R}_{++} \rangle &= -\Gamma \langle R_{++} \rangle + \frac{\Gamma}{2}, \\
\langle \dot{R}_{--} \rangle &= -\Gamma \langle R_{--} \rangle + \frac{\Gamma}{2}, \\
\langle \dot{R}_{+-} \rangle &= -\left(\Gamma_+ + i\Omega\right) \langle R_{+-} \rangle + \Gamma_- \langle R_{-+} \rangle, \\
\langle \dot{R}_{-+} \rangle &= -\left(\Gamma_+ - i\Omega\right) \langle R_{-+} \rangle + \Gamma_- \langle R_{+-} \rangle,
\end{align*}
\]

where $\Gamma_{\pm} = (\Gamma \pm \gamma_z)/2$, $R_{lk} = |l\rangle \langle k| (l, k = \pm)$ is an atomic downward transition operator between the dressed states $|l\rangle$ and $|k\rangle$ of two near-lying dressed doublets. Eqs. (9) and (10) describe the atomic downward transitions between the two dressed states of two adjacent dressed doublets, whereas, eq. (11) (eq. (12)) represents transitions from the dressed state $|+\rangle$ ($|-\rangle$) of one dressed doublet to the dressed state $|-\rangle$ ($|+\rangle$) of the next dressed doublet. If the Rabi frequency $\Omega$ is much greater than $\Gamma_{\pm}$, the terms associated with different resonant frequencies, $\Gamma_- \langle R_{-+} \rangle$ in eq. (11) and $\Gamma_- \langle R_{+-} \rangle$ in eq. (12) are negligible under the secular approximation \( \Omega \gg \gamma \). Consequently, the both transitions, $|+\rangle \rightarrow |-\rangle$ and $|-\rangle \rightarrow |+\rangle$ are independent. Otherwise, they are correlated \( \Lambda \), i.e., as the atom decays from $|+\rangle$ to $|-\rangle$ it drives the other transition from $|-\rangle$ to $|+\rangle$, and vice versa. This reflects the fact that fluorescent photons emitting from these transitions are indistinguishable so that quantum interference between these transition channels \( \Lambda \) dominates.

It is well known that the resonance fluorescence can be described by spontaneous emissions of the atom downward the ladder of the dressed-state doublet \( \Lambda \). The atomic decays between same dressed states of two adjacent dressed doublets, governed by eqs. (9) and (10), give rise to a spectral component

\[
\Lambda_0(\omega) = \frac{\Gamma}{4(\Gamma^2 + \omega^2)},
\]

which centres at the laser frequency and has a linewidth $2\Gamma$ and a height $1/(4\Gamma)$. The spectrum broadens and is suppressed as the laser linewidth increases.

Whereas, the other transitions, described by eqs. (11) and (12), result in a spectrum

\[
\Lambda_1(\omega) = \frac{1}{4} \text{Re} \left[ \frac{\gamma_z + i\omega}{(\Gamma_+ + i\omega)^2 + \Omega^2 - \Gamma_+^2} \right],
\]

whose position and feature are dependent on the laser linewidth $L$ and intensity $\Omega$.

The total fluorescence spectrum consists of the two spectra, $\Lambda(\omega) = \Lambda_0(\omega) + \Lambda_1(\omega)$, which are demonstrated in Fig. 4 for different laser linewidths. The spectrum $\Lambda_0(\omega)$ always shows a Lorentzian shape located at line centre, which is independent of the laser intensity, but varies with the laser linewidth. As the linewidth increases the spectral height is suppressed and the spectral width is broadened. Whereas, $\Lambda_1(\omega)$ is sensitive to the both parameters. When $L \ll \Omega$, $\Lambda_1(\omega)$ exhibits a well-resolved, two-peak structure, as shown in Fig. 4a. This is because the dressed doublet $|\pm\rangle$ is well separated, and the resultant transitions $|+\rangle \rightarrow |-\rangle$ and $|-\rangle \rightarrow |+\rangle$ have different (distinguished) resonance frequencies $\omega \pm \Omega$. Correspondently, the total fluorescence spectrum has a three-peak Mollow structure. When $L \geq \Omega$, $\Lambda_1(\omega)$ shows a dip bored into a wide bell-shape spectrum \( \Lambda_2 \), as depicted in Figs. 4b–4d. The total spectrum thus has a hole burning profile at line centre. The larger the laser linewidth is, the narrower the hole burning is. It is obvious that the hole burning (dip) profile originates from the correlated transitions (quantum interference) between the dressed states $|\pm\rangle$ in two adjacent dressed doublets.

When the atom-laser detuning is taken into account, the populations in the dressed states $|\pm\rangle$ are not same. Hence, the transitions $|+\rangle \rightarrow |-\rangle$ and $|-\rangle \rightarrow |+\rangle$ have different probability amplitudes. The resultant fluorescence spectrum is asymmetric. The total spectrum thus exhibits a dispersive-like profile at line centre.
In summary, we demonstrate that when a two-level atom is excited with a strong laser field with a broad bandwidth due to phase diffusions, the resonance fluorescence spectrum may exhibit the anomalous, narrow resonance features, such as hole-burning and dispersive profiles at line centre. The physics behind the anomalous spectral features is quantum interference between different dressed-state transition channels. From the experimental point of view, observing these features in the system is much easier than that in a squeezed vacuum [4].

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FIG. 1. The resonance fluorescence spectrum $\Lambda(\omega)$ for $\gamma = 1$, $\Omega = 50\gamma$, $\Delta = 0$, and different laser bandwidths: $L = 10\gamma$ (a), $L = 50\gamma$ (b), $L = 100\gamma$ (c), $L = 200\gamma$ (d).

FIG. 2. Three dimensional fluorescence spectrum $\Lambda(\omega)$ for $\gamma = 1$, $\Omega = 50\gamma$, $\Delta = 0$.

FIG. 3. Same as FIG. 1, but for $\gamma = 1$, $\Omega = 50\gamma$, $L = 200\gamma$, and different laser-atom detunings: $\Delta = 50\gamma$ (a), $\Delta = 100\gamma$ (b), $\Delta = 200\gamma$ (c), $\Delta = 400\gamma$ (d).

FIG. 4. Spectral components, $\Lambda_0(\omega)$ (dotted curve) and $\Lambda_1(\omega)$ (dashed curve), and the total fluorescence spectrum $\Lambda(\omega) = \Lambda_0(\omega) + \Lambda_1(\omega)$ (solid curve), where values of the parameters are set same as those in Fig. 1.
\[ \Lambda_0(\omega), \Lambda_1(\omega), \Lambda(\omega) \]

(a) 

(b) 

(c) 

(d)