Modification of resonance fluorescence and absorption in a Λ system by four-wave mixing

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A universal mechanism destroying coherence in a Λ system driven by two resonant laser fields due to four-photon interactions is analyzed theoretically. It is shown that this mechanism gives rise to novel spectral structures in resonance fluorescence. The ''dark resonance'' in absorption (dispersion) spectra is affected as well. [S1050-2947(98)09411-6]

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Three-level atomic systems compared to their two-level counterparts display a much broader range of new effects as a result of coherence among the states induced by the radiation and quantum interference. Among them the coherent population trapping (CPT) is the most intriguing phenomenon that has been already studied both experimentally and theoretically (see [1] and references therein). It is most conspicuous for the Λ transition between two closely spaced long-lived levels optically coupled to a third distant short-lived level by two continuous coherent radiation fields [Fig. 1(a)]. In absorption spectra, the coherent superposition of closely spaced levels leads to a very narrow dip of induced transparency or, equivalently, a nonabsorbing dark resonance when resonance fluorescence is observed.

Any process destroying the coherence of the lower two levels of the Λ system results in a population n_3 of the upper level and residual fluorescence is proportional to Γ_{12}, the relaxation rate of this coherence, which also determines the width of the dark resonance. Experimentally, relaxation due to collisions and laser jitter can be made negligible, thus the residual population n_3 as well as the width of the resonance should be determined only by the length of the interaction time.

With this Brief Report we show that due to nonlinear processes in the case of equal polarizations of the driving fields the ''darkness'' or ultimate width of the CPT resonance is intrinsically limited by a novel type of sidebands in resonance fluorescence, which are present even in the absence of collisions and laser jitter. These single atom processes are caused by four-wave mixing (FWM) processes, which are significant in the near-resonant case. They are closely related also to FWM discussed in [2] for the case of dense media (see also [1], p. 303).

Let us consider a Λ system, which consists of three electronic levels with transition frequencies $\omega_{13},\omega_{23}\gg\omega_{12}$ [Fig. 1(a)]. Two coherent fields, $E\cos(\omega_L t)+E'\cos(\omega_L' t)$, drive the transitions $1\leftrightarrow 3$ and $2\leftrightarrow 3$, respectively. In the vicinity of the Raman resonance condition, the Λ system's Hamiltonian in the interaction representation takes the form

$$\hat{\mathcal{H}}_\Lambda=\hat{\mathcal{H}}_a+\hat{\mathcal{H}}_I, \tag{1}$$

where $\hat{\mathcal{H}}_a=\hbar\omega_{12}|2\rangle\langle 2|+\hbar\omega_{13}|3\rangle\langle 3|$ is the atomic Hamiltonian (we assume that level $|1\rangle$ has zero energy) and

$$\hat{\mathcal{H}}_I(t)=\hbar[g(|1\rangle\langle 3|+\text{H.c.})+\overline{g}(|2\rangle\langle 3|e^{-i\Delta t}+\text{H.c.})]$$
$$+\hbar[g'(|2\rangle\langle 3|+\text{H.c.})+\overline{g'}(|1\rangle\langle 3|e^{i\Delta t}+\text{H.c.})] \tag{2}$$

is the interaction Hamiltonian with the bichromatic detuning $\Delta=\omega_L-\omega_L'\approx\omega_{12}$. The coupling strengths, i.e., the Rabi frequencies, depend as usual on the amplitudes E,E' of the incident fields and the dipole matrix elements d_{13}, d_{23}: $g=\vec{d}_{13}\cdot\vec{E}$, $g'=\vec{d}_{23}\cdot\vec{E}'$, $\overline{g}=\vec{d}_{23}\cdot\vec{E}$, and $\overline{g'}=\vec{d}_{13}\cdot\vec{E}'$. Polarizations of the driving fields, which are explicitly taken into account in the coupling strengths, are considered the same.

In the case of interest, when ω_L and ω_L' are close to ω_{13} and ω_{23}, respectively, we can rewrite $\hat{\mathcal{H}}_a$ in the form

$$\hat{\mathcal{H}}_a=\hat{\mathcal{H}}_0+\hat{\mathcal{H}}_\delta$$

with the ''unperturbed'' and ''perturbing'' Hamiltonians

$$\hat{\mathcal{H}}_0=\hbar(\omega_L|3\rangle\langle 3|-\Delta|2\rangle\langle 2|),$$

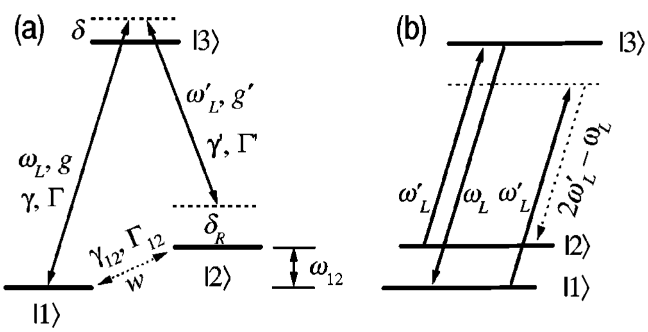

FIG. 1. (a) A driven Λ system: γ, γ', and γ_{12} are the radiation decay rates, Γ, Γ', and Γ_{12} are the dephasing rates, and w is the incoherent pumping rate of the ground levels subsystem. (b) A four-photon process due to FWM in the Λ system at the resulting frequency $2\omega_L'-\omega_L$. Another one (not shown here) takes place at the frequency $2\omega_L-\omega_L'$.

$$\hat{\mathcal{H}}_\delta = -\hbar\delta|3\rangle\langle 3| + \hbar\delta_R|2\rangle\langle 2|,$$

correspondingly. Here $\delta=\omega_L-\omega_{13}$ is the one-photon detuning for the $|1\rangle\rightarrow|3\rangle$ transition and $\delta_R=\omega_L-\omega_L'-\omega_{12}$ is the two-photon Raman detuning [Fig. 1(a)], respectively.

The dynamics of the Λ system with the Hamiltonian (1) is governed by the equation

$$\frac{d\hat{A}}{dt}=\frac{i}{\hbar}[\hat{\mathcal{H}}_0+\hat{\mathcal{H}}_\delta+\hat{\mathcal{H}}_I(t),\hat{A}]+\mathcal{L}_r\hat{A}$$
$$=[\mathcal{L}_0+\mathcal{L}_\delta+\mathcal{L}_I(t)+\mathcal{L}_r]\hat{A}=\mathcal{L}\hat{A} \quad (3)$$

for the operator $\hat{A}$ of an observable A. Here $\mathcal{L}_0$, $\mathcal{L}_\delta$, and $\mathcal{L}_I(t)$ represent the dynamics due to the corresponding Hamiltonians $\hat{\mathcal{H}}_0$, $\hat{\mathcal{H}}_\delta$, and $\hat{\mathcal{H}}_I(t)$ and $\mathcal{L}_r$ describes the relaxation in the system, which we assume to be Markovian. We define $\mathcal{L}_r$ in the basis of nine operators $\hat{e}_j=|k\rangle\langle k|$, $(|k\rangle\langle l|+|l\rangle\langle k|)/\sqrt{2}$, $i(|k\rangle\langle l|-|l\rangle\langle k|)/\sqrt{2}$ $(j=1,\ldots,9;\ k,l=1,2,3;\ k\neq l)$ with a set of phenomenological parameters shown in Fig. 1(a).

The dynamics of the driven Λ system is described by Eqs. (2) and (3) both within the rotation wave approximation (RWA) and beyond it. The latter is necessary to account for the FWM processes shown in Fig. 1(b), which are significant in the strong field limit. We will now examine how the FWM processes affect both the resonance fluorescence and absorption spectra of the Λ system.

Let us first consider the *resonance fluorescence* in terms of a simple dressed atom model [3,4] and then provide both an analytical solution and computer simulation results for the fluorescence spectra.

After introducing a coupled quantum state, $|c\rangle=g_\Lambda^{-1}(g|1\rangle+g'|2\rangle)$, which is coupled to the excited level $|3\rangle$ with an effective rate $g_\Lambda=\sqrt{g^2+g'^2}$, and an orthogonal, noncoupled quantum state, $|n\rangle=-g_\Lambda^{-1}(g'|1\rangle-g|2\rangle)$, the Hamiltonian (1) can be readily decomposed for $\delta_R=0$ into an effective two-level system of $|c\rangle$ and $|3\rangle$ states and noncoupled state $|n\rangle$, not coupled to the $|c\rangle\oplus|3\rangle$ system [1]. Relaxation in the system leads to the depopulation of the $|c\rangle\oplus|3\rangle$ system through spontaneous decay into the noncoupled state, which is coupled to $|c\rangle$ through the spontaneous decay rate γ_{12}, elastic dephasings rate Γ_{12}, incoherent pumping rate w of the transition $|1\rangle\rightarrow|2\rangle$, and Raman detuning δ_R. These parameters determine how perfect is the dark or CPT state.

In the dressed atom model bare states $|1\rangle$, $|2\rangle$, and $|3\rangle$ are replaced by manifolds of dressed states shown in Fig. 2(a) (we accepted the notations of [3,5]). In the absence of atomic interaction with the light field, these manifolds are degenerate since resonant absorption of a photon from either light field brings the system into the excited state. At perfect CPT condition $(\delta,\delta_R=0)$, the positions of the spectral lines can be directly derived from an analysis of allowed spontaneous decay channels [Fig. 2(a), inset]. Five incoherent lines for the two structures centered at the laser frequencies ω_L,ω_L' (inner and outer satellites are shifted to $\pm g_\Lambda/2$ and $\pm g_\Lambda$, respectively) are known from an earlier theoretical analysis [3,4]. They will be called RWA multiplets. In addition, our model predicts two new structures at the four-photon frequencies $2\omega_L-\omega_L'$, $2\omega_L'-\omega_L$ (shifted to $\pm g_\Lambda/2$) which are caused by FWM processes and will be called FWM multiplets. A complete fluorescence spectrum at the CPT condition shown in Fig. 2(b) includes also the coherent lines (centered at the laser and four-photon frequencies), which may also be present in the spectrum. In addition to the fluorescence spectrum at optical frequencies, one could register also a spectrum of oscillations in the rf range at the frequency ω_{12}, which has the same power spectrum as the FWM spectrum.

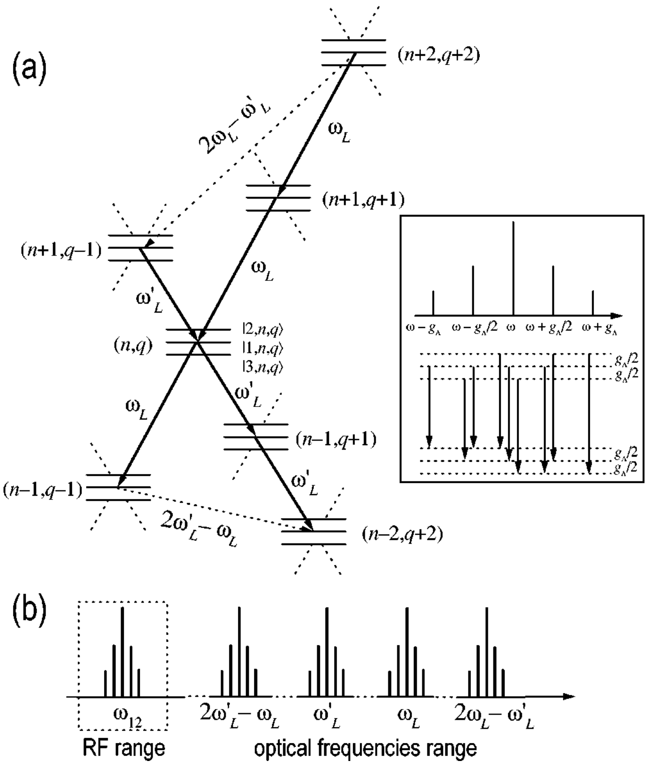

FIG. 2. (a) Dressed energy level diagram for the driven Λ system. The manifold (n,q) is separated from the manifolds $(n\pm1,q\mp1)$ and $(n\pm1,q\mp1)$ by the laser frequencies ω_L and ω_L', respectively. Here $n=n_1+n_2$ and $q=n_1-n_2$ for n_1, n_2 being the mean number of photons in the modes ω_L and ω_L'. Solid arrows show resonant couplings describing emission of photons from $|3\rangle\rightarrow|1\rangle$ or $|3\rangle\rightarrow|2\rangle$ transitions and dotted arrows correspond to the lowest-order nonresonant couplings describing emission of photons due to the four-photon interactions at the frequencies $2\omega_L-\omega_L'$ and $2\omega_L'-\omega_L$. The inset shows possible spontaneous decays (solid arrows) from any two coupled manifolds separated by the frequency ω, which is ω_L or ω_L' for the resonant couplings and $2\omega_L-\omega_L'$ or $2\omega_L'-\omega_L$ for the nonresonant ones. In the case of resonant driving laser fields shown here, the perturbed states of any manifold are separated by $g_\Lambda/2$ and the resulting positions of the lines in the fluorescence spectrum are shown above. (b) Positions of lines in the fluorescence spectrum of the resonantly driven Λ system. In the dotted box, the positions of lines in the power spectrum of the rf vibrations at the frequency ω_{12} accompanying fluorescence are shown.

For an analysis of the line shapes in the fluorescence spectrum and their intensities, it is necessary to explicitly calculate the spectrum. Under the conventional assumption of a Markovian character of atomic fluctuations, the two-time correlation function describing atomic radiation takes in the

Heisenberg representation the form (for a detailed technique, see [6])

$$\mathcal{K}(\tau)=\langle\hat{\rho}_0 S(0,t)|\hat{\sigma}^-(t)[S(t,t+\tau)\hat{\sigma}^+(t+\tau)]\rangle, \quad (4)$$

where $\hat{\sigma}^{\pm}(t)$ are the positive and negative frequency Heisenberg atomic transition operators. $S(0,t)$ and $S(t,t+\tau)$ are the evolution superoperator at the time intervals $(0,t)$ and $(t,t+\tau)$, respectively, and $\hat{\rho}_0 S(0,t)$ is the density matrix $\hat{\rho}(t)$ at time t. The spectrum is calculated then by Fourier transform of Eq. (4).

The evolution superoperator $S(0,t)$ corresponding to the Liouvillian in Eq. (3) can be expanded to the first order as

$$S(0,t)=S_{\mathrm{RWA}}(0,t)\left[1+\int_0^t \delta\mathcal{L}_I(\tau)d\tau\right]e^{\mathcal{L}_0 t}, \quad (5)$$

where $S_{\mathrm{RWA}}(0,t)=\exp(\mathcal{L}_{\mathrm{RWA}}t)$ with $\mathcal{L}_{\mathrm{RWA}}=\mathcal{L}_\delta+\mathcal{L}_r+\langle\mathcal{L}_I\rangle_t$ describes the dynamics of the system within the RWA and

$$\delta\mathcal{L}_I(t)=e^{\mathcal{L}_0 t}\mathcal{L}_I(t)e^{-\mathcal{L}_0 t}-\langle\mathcal{L}_I\rangle_t \quad (6)$$

is the deviation of the laser excitation superoperator from its time-averaged value oscillating with the laser frequencies and their combinations. Equation (5) extends beyond the RWA describing dynamics due to the FWM processes. A simple analysis of Eq. (6) shows that it contains terms oscillating at the biharmonic detuning Δ and, as a result, additional new spectral components appear at the frequencies $\omega_L-\Delta=2\omega_L-\omega'_L$ and $\omega'_L+\Delta=2\omega'_L-\omega_L$. It is known that FWM leads to the generation of coherent Stokes and anti-Stokes waves (CSRS and CARS) at these frequencies [7]. In our result, however, these nonlinear resonances are accompanied by Mollow-type sidebands resulting from incoherent scattering processes involving four photons.

To elucidate the nature of the fluorescence spectrum, we have performed both computer simulations of Eq. (4) for the general case (Fig. 3) and derived an analytical solution, which is presented for the case of resonant excitation ($\delta,\delta_R=0$) and $w,\gamma_{12},\Gamma_{12}\ll\gamma$ below. Spectral densities for the resonant RWA multiplets, $\mathcal{F}_{\mathrm{RWA}}$, at the laser frequencies ω_L,ω'_L and for the FWM multiplets, $\mathcal{F}_{\mathrm{FWM}}$, at the frequencies $2\omega_L-\omega'_L, 2\omega'_L-\omega_L$ have asymptotically with respect to two independent parameters $\gamma/g_\Lambda\to 0$ and $g_\Lambda/\omega_{12}\to 0$ the form (in photons/Hz)

$$\begin{aligned}\mathcal{F}_{\mathrm{RWA}}=&(\tilde{\Gamma}_{12}/4\gamma)\{L[0,(\tilde{\Gamma}_{13}+\tilde{\Gamma}_{23})/2]\\&+L[\pm\tilde{g}_\Lambda/2,(\tilde{\Gamma}_{13}+\tilde{\Gamma}_{23})/4]\\&+(1/2)L[\pm\tilde{g}_\Lambda,(3+\tilde{\Gamma}_{13}+\tilde{\Gamma}_{23})/4]\},\end{aligned} \quad (7)$$

$$\begin{aligned}\mathcal{F}_{\mathrm{FWM}}=&(g_\Lambda^2/16\gamma\omega_{12}^2)\{L[\pm\tilde{g}_\Lambda/2,(\tilde{\Gamma}_{13}+\tilde{\Gamma}_{23})/4]\\&+(\tilde{\Gamma}_{12}/4)[18L[0,1/2]\\&+L[\pm\tilde{g}_\Lambda,(3+\tilde{\Gamma}_{13}+\tilde{\Gamma}_{23})/4]]\},\end{aligned} \quad (8)$$

where $L(\tilde{g},\tilde{\Gamma})=\tilde{\Gamma}/[(\omega/\gamma-\tilde{g})^2+\tilde{\Gamma}^2]$ is the Lorentzian function and a tilde denotes parameters normalized to γ. Equations (7) and (8) describe only inelastic scattering components in the fluorescence spectrum. There are also coherent lines due to elastic scattering in the FWM spectrum at the frequencies $2\omega_L-\omega'_L, 2\omega'_L-\omega_L$, but the RWA spectrum contains no coherent lines.

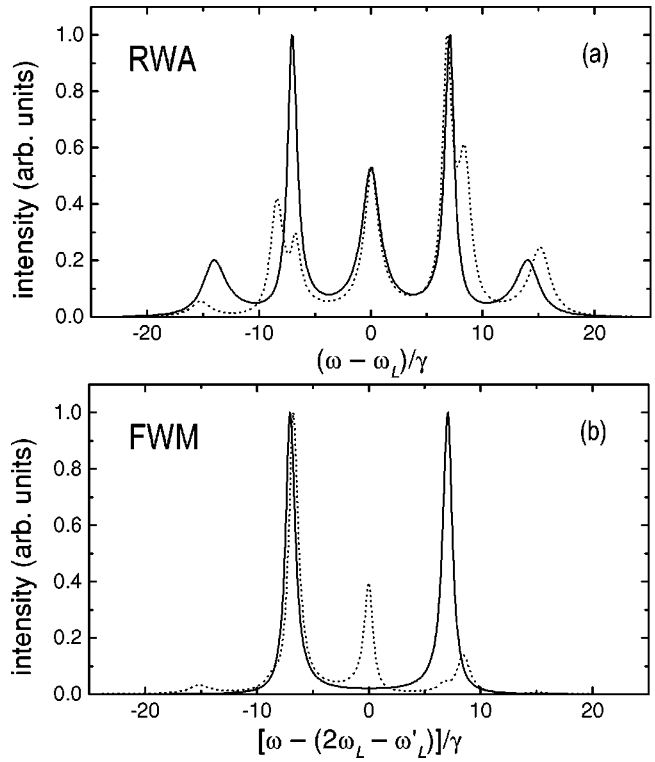

FIG. 3. Calculated resonance fluorescence spectra from the driven symmetric Λ system at the resonant ($\delta,\delta_R=0$, solid curves) and off-resonant ($\delta=0$, $\delta_R=5$, dotted curves) excitation with the equal driving field intensities ($g_1=g_2=10$). Shown spectra correspond to the RWA (a) and FWM (b) multiplets (coherent line, RWA multiplet at the ω'_L frequency, and FWM multiplet at the $2\omega_L-\omega'_L$ frequency are not shown). The parameters used in the calculations are as follows: $\tilde{\gamma}_{13}=\tilde{\gamma}_{23}=1$, $\tilde{\gamma}_{12}=0$, $\tilde{\Gamma}_{12}=10^{-4}$.

For perfect CPT conditions ($\delta,\delta_R=0$) and for $\gamma_{12}=0,\Gamma_{12}\to 0$, according to Eqs. (7) and (8), the RWA spectrum consists of five incoherent lines, whereas in the FWM spectrum both the central incoherent line and the sidebands at $\pm g_\Lambda$ vanish with $\Gamma_{12}\to 0$, resulting in a spectrum which has a central coherent line and two incoherent satellites at $\pm g_\Lambda/2$ [Fig. 3, solid curves; see also Fig. 2(a)].

For $\delta_R\neq 0$, both the RWA and FWM spectra are enriched—up to seven lines appear in the RWA multiplets and up to five lines in the FWM ones (Fig. 3, dotted curves). The RWA multiplets in the fluorescence spectrum of a bichromatically excited Λ system received above were discussed for the first time in [3], then in [4,5], and recently were investigated experimentally for a trapped and cooled Ba^+ ion in [8], whereas the FWM multiplets we discuss here make a novel result. These spectra can be registered experimentally, for example, from an atomic beam of Cs atoms traversing a spectrum analyzer (e.g., a 5-mm Fabry-Pérot cavity with the finesse equal to 10^4) with orthogonal excitation by laser light [9]. For an atomic beam with 10^9 atoms $s^{-1}\,mm^{-2}$ and a saturation intensity of 1.1 mW cm^{-2}, a coupling factor of $g_\Lambda=10^2\gamma$ is readily obtained from 30 mW of laser light at 852 nm focused to 1 mm spot size.

Under these moderate conditions one estimates that photon counting should detect more than 10^3 photons s^{-1}, which is well within the realm of conventional photon counters.

One of the most important consequences is, in our view, that according to Eq. (8) for constant intensity the RWA multiplets completely vanish for $\Gamma_{12}\to 0$, whereas the FWM spectra survive. They are caused by spontaneous decay of the excited state to the mixture of the coupled and noncoupled states due to the FWM processes. As a result, an additional FWM contribution to the dephasing rate Γ_{12} of the ground levels appears, which reads [6]

$$\Gamma_{12}^{\mathrm{FWM}} \approx \frac{g_\Lambda^2}{4\omega_{12}^2}\frac{\gamma}{2} \tag{9}$$

and for Cs atoms ($^2S_{1/2}\to {}^2P_{3/2}$ transition) at $I\sim 1$ W/cm^2 has a value $\Gamma_{12}^{\mathrm{FWM}}\sim 10^4$ s^{-1}, which is large compared to Γ_{12}. At the same time, it is well known that conventional power broadening plays a key role at low and moderate intensities [1]. Therefore, the complete dephasing rate reads

$$(\Gamma_{12}^{\mathrm{total}})^2 = (\Gamma_{12}^{\mathrm{power}})^2 + (\Gamma_{12}^{\mathrm{FWM}})^2 = \left(\Gamma_{12}^2 + \Gamma_{12}\frac{g_\Lambda^2}{2\gamma}\right) + \frac{g_\Lambda^4}{64\omega_{12}^4}\gamma^2.$$

Experimentally, the influence of collisions and laser jitter, as well as Doppler broadening, can be made negligible [8]. We therefore can neglect Γ_{12} and, as a result, $\Gamma_{12}^{\mathrm{power}}$, but not $\Gamma_{12}^{\mathrm{FWM}}$, which dominates at large laser intensity. In other words, we can say that it sets a *fundamental limit* to the dephasing rate of the ground subsystem and thus to the darkness of the CPT resonance.

The FWM processes affect also the *refractive index* of the Λ media [1,2], which in simplified form reads

$$n_k' + i n_k'' = 1 + 0.0289 p \lambda^3 \frac{n_3}{\tilde g_k^2}\left[\frac{(\tilde g^2 - \tilde g'^2)\tilde\Gamma_{12}\tilde\delta + \tilde g_\Lambda^2 \tilde\delta_{\mathrm R}}{\tilde g_\Lambda^2 \tilde\Gamma_{12} + 4\tilde\Gamma_{12}^2 + 4\tilde\delta_{\mathrm R}^2} - \tilde\delta + i\right], \tag{10}$$

where $k=1,2$ denotes values for the respective laser fields with the frequencies ω_L $(k=1)$ and ω_L' $(k=2)$, and the average population $n_3 = \langle 0|\hat n_3\rangle$ is given by

$$n_3^{-1} = 3 + \frac{2\tilde g_\Lambda^2}{\tilde\Gamma \tilde g^2 \tilde g'^2}(\tilde\Gamma^2 + \tilde\delta^2) + \frac{\tilde g_\Lambda^4 \tilde\Gamma^2 \tilde\Gamma_{12} + \tilde g_\Lambda^6 \tilde\Gamma/4 - (\tilde g'^2 - \tilde g^2)^2 \tilde\Gamma_{12}\tilde\delta^2 + 2\tilde\Gamma \tilde g_\Lambda^2 (\tilde g'^2 - \tilde g^2)\tilde\delta_R \tilde\delta}{2\tilde\Gamma^2 \tilde g^2 \tilde g'^2 (\tilde\delta_R^2 + \tilde\Gamma_{12}^2 + \tilde\Gamma_{12}\tilde g_\Lambda^2/4\tilde\Gamma)}. \tag{11}$$

Our calculations for the dark resonance in absorption spectra show that the FWM reduces the dip of the dark resonance in an absorption spectrum of a vapor of Cs atoms insignificantly, but in the case of K_{41} vapor it leads to a strong vanishing of the resonance [6]. This essential difference could be easily understood from Eq. (9) for the $\Gamma_{12}^{\mathrm{FWM}}$, which has the frequency splitting ω_{12} in the denominator. For K_{41}, this parameter is much smaller than for Cs, with the rest of the parameters of the model being the same order of magnitude, and therefore determines the FWM effect much stronger in K_{41} than in Cs. One should keep this in mind while considering different applications of the dark resonances. For magnetometry [1], for instance, our results indicate that the use of Cs atoms is preferable.

Experimentally, the importance of the FWM mechanism for high-resolution spectroscopy in dense coherent Λ media was demonstrated in [2] (see also references therein for related works). Our result, however, shows that the FWM nonlinear processes are peculiar to even a single atom.

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[1] E. Arimondo, in *Progress in Optics*, edited by E. Wolf (Elsevier, Amsterdam, 1996), Vol. 35, p. 257.
[2] M. D. Lukin *et al.*, Phys. Rev. Lett. **79**, 2959 (1997).
[3] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977); **10**, 2311 (1977).
[4] L. M. Narducci *et al.*, Phys. Rev. A **42**, 1630 (1990); A. S. Manka *et al.*, *ibid.* **43**, 3748 (1991).
[5] M. R. Ferguson, Z. Ficek, and B. J. Dalton, Phys. Rev. A **54**, 2379 (1996).
[6] B. A. Grishanin, V. N. Zadkov, and D. Meschede, Zh. Eksp. Teor. Fiz. **113**, 144 (1998) [JETP **113**, 144 (1998)].
[7] R. Y. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).
[8] Y. Stalgies *et al.*, Europhys. Lett. **35**, 259 (1996).
[9] D. J. Gauthier, Y. Zhu, and T. W. Mossberg, Phys. Rev. Lett. **66**, 2460 (1991).