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Elimination of Doppler broadening at coherently driven quantum transitions

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Possibilities of elimination of Doppler broadening by making use of atomic coherence in ladder- and folded-type configurations of quantum transitions are considered. A simple physical analysis in terms of modification of the frequency-correlation factor for cascade and multiphoton processes in strong driving fields is developed. [S1050-2947(99)50402-2]

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The possibilities for controlling material optical properties through the quantum coherence have attracted much attention in recent years (for a review, see [1,2]). Among them are the formation of spectral ranges of amplification without inversion (AWI) [3] and transparency [4], increased dispersion [5], enhanced nonlinear optical cross sections [6], and population transfer to highly excited states [7]. The fact that quantum interference in coherently driven atom gives rise to differences in the spectral properties of the emission and absorption probabilities and, therefore, to the AWI was demonstrated in an early paper [8]. The possibilities for manipulating the optical properties of one transition with the controlling field at the adjacent transition of a three-level atom and, in particular, conditions for AWI were first investigated in [9]. (Some other early publications are reviewed in [2,10].) A particular case of AWI with the continuum states involved was considered in Ref. [11]. There are relatively few publications considering the effect of Doppler broadening in coherent coupling and various manifestations of the nonlinear interference effects [8,10,12–15]. A possible sub-Doppler resolution using intense control fields was recently considered in [16].

Inhomogeneous Doppler broadening of atomic and molecular transitions in gases decreases the number of atoms coherently driven by laser fields [10,17]. A widely used method of elimination of the Doppler broadening under two-photon excitation in the field of a standing wave is restricted to ladder-type transitions. A large detuning from the intermediate resonance that decreases the process's cross section is characteristic of this type of elimination of the resonance inhomogeneous broadening [17]. In an early paper [12] and in consequent publications [13,14] (for a review, see [10]), methods overcoming these limitations for coherently driven three-level atoms were proposed; these methods are based on the use of strong driving fields and a change of the frequency-correlation properties of resonant multiphoton processes in strong resonance fields. It was shown that elimination of inhomogeneous broadening is possible even for different frequencies of the coupled fields, such as those in Raman-type electron transitions. These methods, however, are typically accompanied by a change of the population of atom levels that significantly complicates the manifestation of coherence effects and their interpretation.

In this paper we propose schemes for the cancellation of Doppler broadening that can be used at quasiresonance with an intermediate level, i.e., in general, for different frequencies of the applied fields. It is important that the proposed schemes not involve a change of the atom level populations and allow observation of atomic coherence Doppler-free effects that are not obscured by the saturation. The fact that the Doppler-broadening elimination occurs at near resonance with an intermediate level dramatically increases the process efficiency and makes possible the observation of such effects in gases at moderate intensities. We also show that using two strong driving fields E_2 and E_3 [see Figs. 1(a) and 2(a)] offers additional ways of eliminating Doppler broadening in the absorption contour of a probe field E_1.

Consider an atom coupled to a strong field E_2 with frequency ω_2 that is close to the transition frequency ω_{21} [see Fig. 1(a), where the field E_3 is first assumed to be off]. Because of the stimulated transitions, the atom oscillates between the upper and lower resonance states; as a result, the corresponding probability amplitudes are modulated. At large frequency of the induced transitions, this fact exhibits itself in the absorption of a probe radiation interacting with an adjacent transition so that the shared energy level splits into two quasilevels (Autler-Townes splitting, ac Stark shifts). The two resonant detunings for the weak probe radiation $\Omega_1^{\pm}=\omega_1^{\pm}-\omega_{10}$ are given by (see, e.g., [12])

$$\Omega_1^{\pm}=-\tfrac{1}{2}\{\Omega_2\pm\sqrt{4|G_2|^2+\Omega_2^2}\}, \tag{1}$$

where $\Omega_2=\omega_2-\omega_{21}$ and $G_2=E_2d_{12}/2\hbar$ is the coupling Rabi frequency.

The correlation factor in the absorption of frequencies ω_1 and ω_2 is defined as [10,12,14]:

$$M_{\pm}=\left|\frac{d\Omega_1^{\pm}}{d\Omega_2}\right|=\frac{1}{2}\left[1\pm\frac{\Omega_2}{\sqrt{4|G_2|^2+\Omega_2^2}}\right]. \tag{2}$$

This factor can be thought of as a "memory factor." It reflects the correlation between the acts of absorption of photons $\hbar\omega_1$ and $\hbar\omega_2$. To illustrate this, we first consider the limit of weak fields ($|G_2|^2\ll\Omega_2^2$), for which we obtain from Eqs. (1) and (2) $\Omega_1^-=0$ and $\Omega_1^+=-\Omega_2$, with the correlation factors equal to $M_-=0$ and $M_+=1$, respectively. Then there exist two quantum pathways. In the first one, after the

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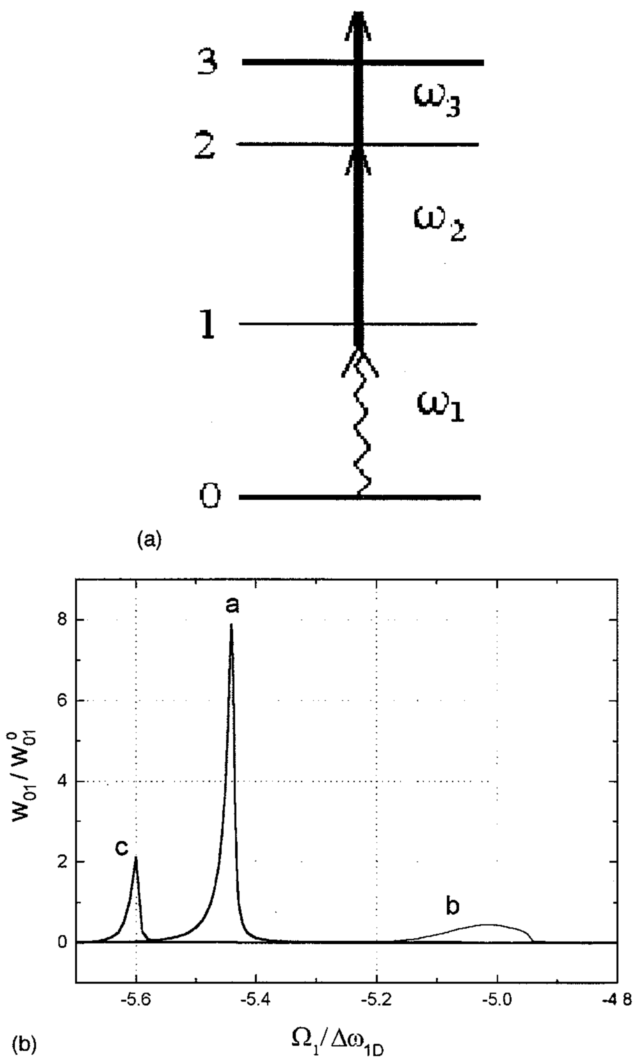

FIG. 1. (a) Ladder-type energy-level configuration. (b) Absorption coefficient for a probe field at ω_1, normalized by its resonant value at $E_2=0$ in a ladder scheme. Curve a, Doppler-free resonance in the presence of a strong counterpropagating field at ω_2 ($E_3=0$). Curve b, same as a, but the conditions of Doppler-broadening elimination are not fulfilled. Curve c illustrates the compensation of the residual Doppler broadening with the aid of the additional strong field E_3.

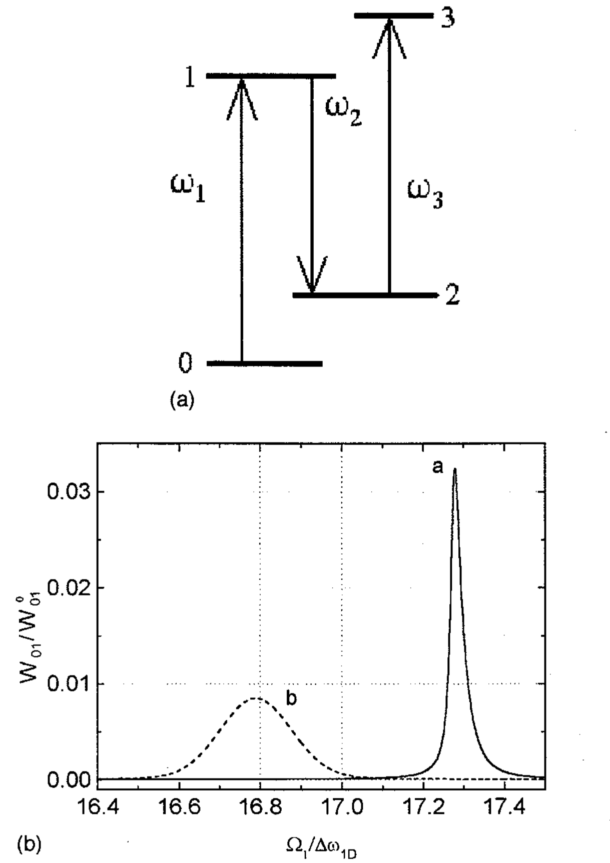

FIG. 2. (a) Raman Λ-type energy-level configurations. (b) Power-induced sub-Doppler resonances in the Raman-type scheme. Curves a and b correspond to the cases where the field E_3 is on and off, respectively.

absorption of photon $\hbar\omega_1$, the atom transfers to the excited intermediate state, resides there within the lifetime Γ_1^{-1}, and then absorbs photon $\hbar\omega_2$, with the absorption probability peaked at the resonant frequency of the adjacent transition ω_{21}. In this case, the atom "forgets" about the origin of the excitation, there is no correlation between the two absorbed photons, and the "memory factor" equals zero. Another pathway is when $\hbar\omega_1$ is absorbed and the atom only virtually appears in the excited state (in the quasilevel shifted by $\hbar\Omega_2$) within the time interval about $|\Omega_1|^{-1}\ll\Gamma_1^{-1}$. Because of that it still keeps the memory about the exciting photon when it absorbs photon $\hbar\omega_2$. As a consequence, the largest probability of the absorption falls at frequency $\omega_2=\omega_{20}-\omega_1$. In this case, there is full frequency correlation for the two quantum transitions and the "memory factor" equals 1. The two above processes correspond to a cascade and two-photon transitions, respectively.

In a similar way, one can consider a Raman-type configuration [see Fig. 2(a), where E_3 is off, for now]. In this case photon $\hbar\omega_2$ is emitted with the highest probability at $\omega_2=\omega_{12}$, for a cascade process ($M=0$), and at $\omega_2=\omega_1-\omega_{20}$, for a two-photon process ($M=1$).

Provided the applied frequencies are close to the resonance with the intermediate level, two coherently driven quasilevels are closely spaced and the two paths considered above interfere. As was first outlined in [8–10], this results in difference in the spectral properties of pure emission and absorption and, as a consequence, leads to the AWI.

With the increase of the Rabi frequency of the driving field, the splitting between the quasilevels increases as well. As a result, each of the $M_\pm$ factors associated with the two quasilevels changes. In this case, redistribution of the correlation ("memory") between the two quantum paths occurs, whereas the total "memory" is conserved, $M_+ + M_- = 1$. In the limit of strong fields ($|G_2|^2\gg\Omega_2^2$), $M_+\approx M_-\approx 1/2$, i.e., the two quantum paths correspond to neither purely two-photon nor cascade processes. The process of "memory" redistribution is accompanied by a modification of the absorption and emission spectral profiles, which enables us to manipulate the spectral absorption-gain line shape as a whole.

Now we take into account the motion of atoms in gases. Because of the Doppler effect for atoms moving with velocity $\mathbf{v}$, all the detunings should be replaced by $\Omega_{1,2}'\equiv\Omega_{1,2}-\mathbf{k}_{1,2}\cdot\mathbf{v}$, where $\mathbf{k}_{1,2}$ are the field wave vectors. From the condition $\Omega_1'=\Omega_1^{\mp}(\Omega_2')$ with the aid of Eq. (1) in the limit $|\Omega_2|\gg|\mathbf{k}_2\cdot\mathbf{v}|$, we obtain the following expansion up to the first order of $|\mathbf{k}_2\cdot\mathbf{v}/\Omega_2|\ll 1$:

$$\Omega_1-\mathbf{k}_1\cdot\mathbf{v}=\Omega_1^{\mp}(\mathbf{k}_2\cdot\mathbf{v}=0)+M_{\pm}\mathbf{k}_2\cdot\mathbf{v}. \tag{3}$$

As follows from Eqs. (1)–(3), the resonance can simultaneously occur for all velocities (and thus inhomogeneous Doppler broadening is eliminated) at $\mathbf{k}_1=-M_{\pm}\mathbf{k}_2$. Another form of this equation is $\mathbf{k}_2=-\mathbf{k}_1(1+|G_2|^2/\Omega_1^2)$. These relations correspond to the compensation of Doppler shifts by the velocity-dependent light-induced resonance shifts [12–14]. For weak fields ($|G_2|^2\ll\Omega_2^2$) Eq. (3) turns to the well-known condition of the two-photon resonance for all atoms independent of their velocities: $\mathbf{k}_1=-\mathbf{k}_2$ ($\omega_1=\omega_2$). For Raman-like (Λ- and V-type) configurations, Eq. (3) acquires the form $\Omega_1-\mathbf{k}_1\cdot\mathbf{v}=-[\Omega_1^{\mp}(\mathbf{k}_2\cdot\mathbf{v}=0)+M_{\pm}\mathbf{k}_2\cdot\mathbf{v}]$.

If the inhomogeneous Doppler widths are much larger than the homogeneous ones (taking into account the power broadening), the spectral lines are described by the Gaussian function with the widths

$$q_{\pm}\bar{v}=\sqrt{(k_1+M_{\pm}k_2)^2-4M_{\pm}k_1k_2\sin^2(\theta/2)}\bar{v},$$

where $\bar{v}$ is the thermal velocity, and θ is the angle between the wave vectors. Provided the value $q_{\pm}\bar{v}$ becomes less than a homogeneous width, the shapes of the sub-Doppler resonances are described by the Lorentzian function with the width $\tilde{\Gamma}_{02}$ (see below).

In a strong driving field, the requirement (3) and its counterpart for the Λ and V configurations can be met at $k_1\neq k_2$ because of the significant difference $M_{\pm}$ from 1. Thus, at the correct choice of intensity of a driving field the considered effects give rise to the resonant coupling of all or most of the atoms in a wide velocity interval. It is important that Doppler cancellation become possible at $\omega_1\neq\omega_2$; in particular, at near resonance with the intermediate state, and even for Λ- and V-type schemes [10,12–14].

Note that if the E_1 that couples the ground and intermediate quantum states is relatively weak, the population motion is negligible and the saturation effects are of no importance. The velocity-dependent ac Stark shifts, however, can be significant because of the strong field E_2 (and E_3, see below), so that the sub-Doppler resonances can be realized without the obscuring saturation, even at near resonance with the intermediate state.

Now we consider possibilities for creating and manipulating Doppler-free resonances with two controlling fields E_2 and E_3 for a ladder-type configuration of the energy levels [Fig. 1(a)]. Provided that the field E_1 is so weak that the population change of the atomic levels can be neglected, the formula for the probability of absorption of photons $\hbar\omega_1$ per unit time is given by

$$W(\Omega_1)=2\,\mathrm{Re}\left\{|G_1|^2\frac{P_{02}P_{03}+|G_3|^2}{P_{01}\tilde{P}_{02}P_{03}}\right\}. \tag{4}$$

Here G_i are the Rabi frequencies for the corresponding transitions, $P_{01}=\Gamma_{01}+i(\Omega_1-\mathbf{k}_1\cdot\mathbf{v})$, $P_{02}=\Gamma_{02}+i[\Omega_1+\Omega_2-(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{v}]$, $P_{03}=\Gamma_{03}+i(\Omega_1+\Omega_2+\Omega_3-\mathbf{k}_s\cdot\mathbf{v})$, $\mathbf{k}_s=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3$, and $\tilde{P}_{02}$ describes the field-modified two-photon resonance: $\tilde{P}_{02}=P_{02}+|G_2|^2/P_{01}+|G_3|^2/P_{03}$. Assuming that the detunings of the field frequencies from one-photon resonances are much greater than the Doppler widths of the transitions, we obtain

$$\tilde{P}_{02}=\tilde{\Gamma}_{02}+i\tilde{\Omega}_{02}-i\left\{\left(1+\frac{|G_2|^2}{\Omega_1^2}\right)\mathbf{k}_1+\mathbf{k}_2+\frac{|G_3|^2}{(\Omega_1+\Omega_2+\Omega_3)^2}\mathbf{k}_s\right\}\cdot\mathbf{v}, \tag{5}$$

where $\tilde{\Omega}_{02}$ and $\tilde{\Gamma}_{02}$ are given by

$$\tilde{\Omega}_{02}=\Omega_1+\Omega_2-\frac{|G_2|^2}{\Omega_1}-\frac{|G_3|^2}{\Omega_1+\Omega_2+\Omega_3},$$

$$\tilde{\Gamma}_{02}=\Gamma_{02}+\frac{|G_2|^2}{\Omega_1^2}\Gamma_{01}+\frac{|G_3|^2}{(\Omega_1+\Omega_2+\Omega_3)^2}\Gamma_{03}. \tag{6}$$

The above expressions (5) and (6) show that if, in the absence of the E_3 field, the requirements for compensation of the Doppler broadening of transition 0-2 are not fulfilled, the presence of additional strong driving field E_3 allows one to obtain a Doppler-free resonance in the absorption of a probe field, for various relations between the wave vectors.

For a numerical illustration of the proposed method we consider the Li atom with the transitions $\lambda_{01}=670.78$, $\lambda_{12}=610.36$, and $\lambda_{23}=1091.91$ (all in nanometers), the homogeneous half width at half maximum (HWHM) $\Gamma_{01}=2.85$, $\Gamma_{12}=8.35$, $\Gamma_{23}=6.30$, and $\Gamma_{02}=5.5$ (all in megahertz), and the Doppler HWHM $\Delta\omega_{1D}=1.36$, $\Delta\omega_{2D}=1.50$, and $\Delta\omega_{3D}=0.84$ (all in gigahertz).

First we consider the case where the field E_3 is off and the waves E_1 and E_2 are counterpropagating. The minimum possible HWHM of the two-photon resonance in a probe standing wave ($\mathbf{k}_1=-\mathbf{k}_2$) is 5.5 MHz. By approaching the intermediate resonance we can increase the absorption cross section at $\Omega_2=6.68$ GHz by about five orders of magnitude. At the same time, due to difference in the radiation frequencies, the resonance becomes broadened (HWHM is $\sqrt{\ln 2}|k_1-k_2|\bar{v}=120$ MHz). At $\Omega_2=6.68$ GHz, the condition $k_1=M_+k_2$ is fulfilled at $G_2=2.32$ GHz [Eq. (2)]. The HWHM of the sub-Doppler "quasi-two-photon" resonance, in this case, is brought down to 9 MHz [Fig. 1(b), curve a]. The resonance occurs at $\Omega_2\approx-7.4$ GHz, which is in good agreement with Eq. (1) for Ω_1^+. At $G_2=1.16$ GHz, which is less than the optimal value [Fig. 1(b), curve b], the peak shifts and broadens (HWHM is about 80 MHz). However, it is possible to remove the residual broadening, giving rise to the Doppler-free resonance [Eq. (6)], and to increase the absorption cross section with the aid of a third strong field E_3 that counterpropagates with respect to E_2 [Fig. 1(b), curve c]. Corresponding parameters are $G_3=2.15$ GHz and $\Omega_3=-5.04$ GHz. The HWHM of the peak c centered at $\Omega_1=-7.6$ GHz is about 8 MHz. All plots are normalized by the linear absorption in the center of the Doppler-broadened one-photon transition. [Note that the differences between the HWHM's of the sub-Doppler resonances on plots and those calculated from Eq. (6) are because of the chosen detunings Ω_2 that are comparable to the Doppler HWHM of the transitions.] This example shows that if the required intensity of a strong field is not available, the Doppler broadening can still be eliminated with an auxiliary strong field.

If the strong field ω_2 is in exact resonance with the transition 1-2, the $M_\pm$ factors become both equal to 1/2 [see Eq. (2)]. In this case, one can obtain a doublet of the Doppler-free resonances only for an adjacent transition with the frequency $\omega_{10}=\omega_{21}/2$. The components of the doublet are symmetrical and equally shifted (by $\pm G_{12}$) from the center of the transition 0-1. The Doppler cancellation occurs, provided the Rabi frequency is much greater than the Doppler width of the transition, so that all atoms are almost equally driven by the fields, independently of the atom velocities.

As mentioned above, Doppler broadening can be also eliminated in the folded (Λ and V) schemes [Fig. 2(a)]. Then Ω_2' must be replaced by $-\Omega_2'$ in all the above expressions. In this case, the resonance condition acquires the form $\tilde{\Omega}_{02}' \approx \tilde{\Omega}_{02}-[(1+|G_2|^2/\Omega_1^2)\mathbf{k}_1\cdot\mathbf{v}-(1+|G_1|^2/\Omega_2^2)\mathbf{k}_2\cdot\mathbf{v}]$, where $\tilde{\Omega}_{02}=\Omega_1-|G_2|^2/\Omega_1-(\Omega_2-|G_1|^2/\Omega_2)$. This expression shows that Doppler cancellation can be obtained by an increase of the field E_1; this process, however, is accompanied by the population transfer to the upper levels [10,14]. In contrast, if E_1 is weak enough, while the others, $E_{2,3}$, are strong, only the ground level is populated. In this case, with the field E_3 being off, the absorption spectral line at ω_1 is Doppler broadened. It cannot be narrowed by increasing G_2 because $\omega_1>\omega_2$. However, it is possible to eliminate the Doppler broadening with the aid of a controlling field at ω_3 that propagates against the two other copropagating beams. By substituting Ω_2 and k_2 for $-\Omega_2$ and $-k_2$ in Eqs. (5) and (6), respectively, we see that the absorption resonance can be narrowed by properly adjusting the intensities of the controlling fields, if $(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{k}_s<0$ $(\mathbf{k}_s=\mathbf{k}_1-\mathbf{k}_2+\mathbf{k}_3)$.

A numerical illustration of this method of Doppler cancellation is given for sodium dimers [18] [see Fig. 2(b)], with $\lambda_{01}=661$, $\lambda_{12}=746$, and $\lambda_{23}=514$ (all in nanometers); the homogeneous HWHM $\Gamma_{01}=\Gamma_{12}=22.3$, $\Gamma_{23}=\Gamma_{03}=17.5$, and $\Gamma_{02}=6.4$ (all in megahertz); and the Doppler HWHM $\Delta\omega_{iD}$ that are equal to 0.68, 0.6, and 0.87 GHz, respectively. Curve *a* in Fig. 2(b) shows the resonance, which is enhanced and narrowed with the aid of a strong counterpropagating field E_3, for moderate intensities and large detunings of the driving fields. Here $G_2=0.37$, $G_3=0.85$, $\Omega_2=11.4$, and $\Omega_3=2.63$ (all in gigahertz). The peak is centered at $\Omega_1=11.7$ GHz and the HWHM is about 11 MHz. Curve *b* shows the same resonance when the field E_3 is off. This clearly indicates that by using the additional strong field E_3 one can eliminate Doppler broadening and thus further reduce the resonance spectral width.

In the end we note that, besides absorption, corresponding sub-Doppler structures can be induced in the refraction index. The considered technique of manipulating Doppler-free coupling can be also used for enhancing nonlinear optical phenomena, e.g., four-wave mixing, and for manipulating the velocity distribution of excited states in gas-kinetic processes in Doppler-broadened media.

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