Sub-Doppler absorption resonances induced by strong radiation

A. S. Baev and A. K. Popov[a)]

Institute of Physics, Siberian Branch of the Russian Academy of Sciences, Krasnoyarsk State University; Krasnoyarsk State Technical University, 660036 Krasnoyarsk, Russia

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New possibilities are demonstrated for eliminating uncompensated Doppler broadening in different types of nonlinear optical processes by means of atomic coherence effects in strong electromagnetic fields are demonstrated.
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1. Doppler broadening of resonances imposes a number of fundamental limitations on the selectivity of the interaction of electromagnetic radiation with atomic and molecular systems.[1] Methods of two-photon excitation in counterpropagating waves are widely used to eliminate the inhomogeneous broadening of two-photon transitions. However, these methods are applicable only in the case of stepped configurations of the transitions and equal frequencies of the interacting waves. Appreciable departures from the intermediate resonances will substantially reduce the two-photon interaction cross sections. For overcoming these limitations, methods based on the use of strong fields and the change in the frequency-correlation properties of multiphoton processes in strong resonance fields were proposed in Ref. 2 and in subsequent publications.[3–9] It was shown that inhomogeneous broadening can be eliminated and sub-Doppler resonances can be realized even for transition configurations of the Raman-scattering type. In this case, resonance interaction with all atoms simultaneously, irrespective of their velocities, is possible. Methods of inducing sub-Doppler spectral structures in strong laser fields have been attracting increasing attention in recent years in the context of using quantum coherence effects to manipulate the optical properties of atoms and molecules in order to form large cross sections for nonlinear optical processes, spectral windows of transmission and amplification without population inversion, large dispersion of materials and effective population of high-lying levels (Refs. 10–12 and references cited therein). In the present letter we propose new ways of compensating the Doppler broadening and of capturing atoms into resonance over a wide interval of velocities on account of coherence and strong-field effects. The results are illustrated by numerical examples for one of the transition schemes.

2. The main idea consists of the following. Let an atom interact with one strong field E_2 at a frequency ω_2 close to the transition frequency ω_{21} (Fig. 1, where $E_3=0$). On account of induced transitions between the resonance states, their probability amplitudes

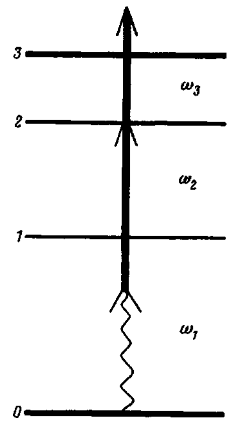

FIG. 1. Transition scheme for eliminating Doppler broadening and capture of atoms into resonance over a wide interval of velocities.

are modulated; for a high frequency of the induced transitions this modulation is manifested, for probe radiation at an adjacent transition, as a splitting of the common level into two quasilevels. Resonance detunings for weak probe radiation $\Omega_1=\omega_1-\omega_{10}$ correspond to the values[2,3,6,9]

$$\Omega_1^{(1,2)}=-\alpha_{1,2}=-\frac{1}{2}\{\Omega_2\pm\sqrt{4|G_{12}|^2+\Omega_2^2}\}, \tag{1}$$

where $\Omega_2=\omega_2-\omega_{21}$, and $G_{12}=E_2d_{12}/2\hbar$ is the Rabi frequency. Hence it follows that the correlation factors of the frequencies ω_1 and ω_2 at the corresponding resonances are determined by the expression

$$M_{1,2}=\frac{d\alpha_{1,2}}{d\Omega_2}=\frac{1}{2}\left[1\pm\frac{\Omega_2}{\sqrt{4|G_{12}|^2+\Omega_2^2}}\right], \tag{2}$$

so that $M_1+M_2=1$ always. In weak fields ($|G_{12}|^2\ll\Omega_2^2$) one has $\Omega_1^{(1)}=-\Omega_2$ and $\Omega_1^{(2)}=0$, while the correlation factors assume the values $M_2=1$ and $M_1=0$, corresponding to two-photon and step processes. In strong fields ($|G_{12}|^2\gg\Omega_2^2$) the correlation factors M_1 and M_2 become equal, with a value of 1/2, i.e., they do not correspond to either single- or two-photon processes.

For atoms moving with velocity $\mathbf{v}$ all the detunings, on account of the Doppler shift, must be replaced by $\Omega'_{1,2}=\Omega_{1,2}-\mathbf{k}_{1,2}\cdot\mathbf{v}$, where $\mathbf{k}_{1,2}$ are the wave vectors of the corresponding radiation. With the aid of Eq. (1), from the condition $\Omega_1^{1,2}-\mathbf{k}_1\cdot\mathbf{v}=-\alpha_{1,2}(\Omega'_2)$ for $|\Omega_2|\gg|\mathbf{k}_2\cdot\mathbf{v}|$ we obtain, to first order in $\mathbf{k}_2\cdot\mathbf{v}/\Omega_2$,

$$\Omega_1^{(1,2)}-\mathbf{k}_1\cdot\mathbf{v}=-(\alpha_{1,2}-M_{1,2}\mathbf{k}_2\cdot\mathbf{v}). \tag{3}$$

In weak fields ($|G_{12}|^2\ll\Omega_2^2$) we obtain from Eq. (3) the well-known condition for two-photon resonance to occur for all atoms simultaneously: $\mathbf{k}_1=-\mathbf{k}_2$ ($\omega_1=\omega_2$). For transition configurations of the Raman-scattering type the condition (3) becomes

$$\Omega_1^{(1,2)}-\mathbf{k}_1\cdot\mathbf{v}=\alpha_{1,2}-M_{1,2}\mathbf{k}_2\cdot\mathbf{v} \tag{4}$$

In strong fields, since $M_{1,2}$ differ substantially from 1, the conditions (3) and (4) can also be satisfied for $k_1 \neq k_2$. Therefore, with the correct choice of the intensity of the strong radiation the field-induced level shifts, which in turn depend on the velocities, can compensate the Doppler shifts and result in capture of the atoms into resonance over the entire velocity interval, even for $\omega_1 \neq \omega_2$, i.e., even at Raman-scattering type transitions.[2,3,9]

3. We shall now present the results of an investigation of new possibilities for manipulating sub-Doppler resonances with the aid of additional strong fields for different types of nonlinear optical resonance processes. For definiteness, we shall consider the nonlinear optical process shown in Fig. 1. For greater clarity, we shall consider the case where the radiation field at the 01 transition is so weak that the change in the populations of the levels can be neglected. Then the formula for the probability of absorption of $\hbar\omega_1$ photons per unit time can be represented in the form (see Ref. 13, Eqs. (24) and (26))

$$w(\Omega_1)=2\,\mathrm{Re}\left\{\frac{|G_{01}|^2}{P_{01}}\,\frac{1}{1+|G_{12}|^2/P_{01}P_{02}[1+|G_{23}|^2/P_{02}P_{03}]}\right\}, \tag{5}$$

where

$$G_{01}=E_1 d_{01}/2\hbar, \quad G_{23}=E_3 d_{23}/2\hbar,$$

$$P_{01}=\Gamma_{01}+i(\Omega_1-\mathbf{k}_1\cdot\mathbf{v}), \quad P_{02}=\Gamma_{02}+i[\Omega_1+\Omega_2-(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{v}],$$

$$P_{03}=\Gamma_{03}+i(\Omega_1+\Omega_2+\Omega_3-\mathbf{k}_\mathbf{s}\cdot\mathbf{v}), \quad \mathbf{k}_\mathbf{s}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3,$$

and Γ_{ij} are the homogeneous half-widths of the corresponding transitions.

Reducing this expression to a common denominator, we obtain

$$w(\Omega_1)=2\,\mathrm{Re}\left\{|G_{01}|^2\,\frac{P_{02}P_{03}+|G_{23}|^2}{P_{01}\tilde{P}_{02}P_{03}}\right\}, \tag{6}$$

where $\tilde{P}_{02}$ describes a two-photon resonance modified by strong fields:

$$\tilde{P}_{02}=P_{02}+\frac{|G_{12}|^2}{P_{01}}+\frac{|G_{23}|^2}{P_{03}}. \tag{7}$$

Assuming that the detuning of the fields from single-photon resonances is much greater than not only the homogeneous but also the Doppler widths of the transitions, and taking the Doppler shifts into account in Eq. (7) in the first nonvanishing approximation, we obtain

$$\tilde{P}_{02}=\tilde{\Gamma}_{02}+i\tilde{\Omega}_{02}-i\left\{\left(1+\frac{|G_{12}|^2}{\Omega_1^2}\right)\mathbf{k}_1+\mathbf{k}_2+\frac{|G_{23}|^2}{(\Omega_1+\Omega_2+\Omega_3)^2}\mathbf{k}_\mathbf{s}\right\}\cdot\mathbf{v}, \tag{8}$$

where $\tilde{\Omega}_{02}$ and $\tilde{\Gamma}_{02}$ have the form

$$\tilde{\Omega}_{02}=\Omega_1+\Omega_2-\frac{|G_{12}|^2}{\Omega_1}-\frac{|G_{23}|^2}{\Omega_1+\Omega_2+\Omega_3}, \tag{9}$$

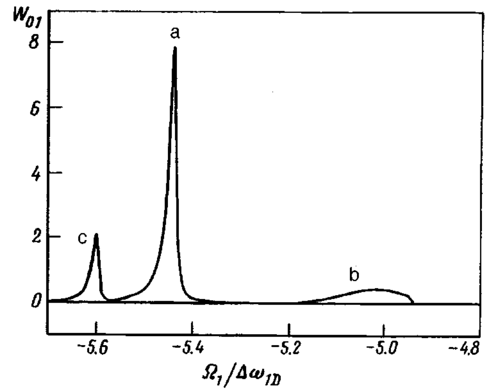

FIG. 2. Probe-field absorption resonances: Curve a — sub-Doppler resonance in a strong field E_2 ($E_3=0$), b — the same, if the conditions for eliminating Doppler broadening are not satisfied, c — compensation of the residual Doppler broadening by means of a strong field E_3. All curves are normalized to the value of the linear absorption at the center of the Doppler-broadened single-photon resonance.

$$\tilde{\Gamma}_{02}=\Gamma_{02}+\frac{|G_{12}|^2}{\Omega_1^2}\Gamma_{01}+\frac{|G_{23}|^2}{(\Omega_1+\Omega_2+\Omega_3)^2}\Gamma_{03}.$$

Formula (8) demonstrates the possibility of obtaining a narrowed two-photon resonance in absorption of the probe field in the presence of two strong fields with different ratios of the wave vectors of the interacting radiations, even if the conditions for compensation of the Doppler broadening at the transition 02 in the absence of the field E_3 are not satisfied. The effects under discussion are a consequence of atomic coherence excited by the strong fields at the interacting transitions.

We shall illustrate the main results in a numerical model of the transitions of the Li atom with the following parameters: $\lambda_{01}=670.784$ nm, $\lambda_{12}=610.364$ nm, $\lambda_{23}=1091.91$ nm, $\Gamma_{01}=2.85$ MHz, $\Gamma_{12}=8.35$ MHz, $\Gamma_{23}=6.30$ MHz, and the Doppler half-widths are, respectively, $\Delta\omega_{1D}=1.362$ GHz, $\Delta\omega_{2D}=1.497$ GHz, and $\Delta\omega_{3D}=0.837$ GHz. The homogeneous half-width of the two-photon transition is $\Gamma_{02}=5.5$ MHz.

4. Let us consider first the case where the field E_3 is switched off. Let the detuning of the field E_2 from resonance be much greater than the Doppler width of the transition 12, and let the intensity correspond to the condition $k_1=M_1k_2$, where M_1 is the correlation factor (2). For the present numerical model with detuning $\Omega_2=6.68$ GHz the Rabi frequency required to eliminate its broadening is $G_{12}=2.32$ GHz. The half-width of the narrowed ''quasi-two-photon'' resonance in the field of the counterpropagating waves of different frequency is equal to approximately 10 MHz, while the position of the resonance is shifted relative to that of the resonance in weak fields (Fig. 2, curve a). The resonance detuning of the probe field in this case is determined by Eq. (1) for $\Omega_1^{(1)}$.

For transitions with frequencies differing by almost a factor of 2 ($\omega_{10}/\omega_{21}\geqslant 0.5$), the quantity G_{12} required to obtain a narrow resonance increases to 10–100 GHz, which corresponds to a light field with intensity of the order of 10 MW/cm^2.

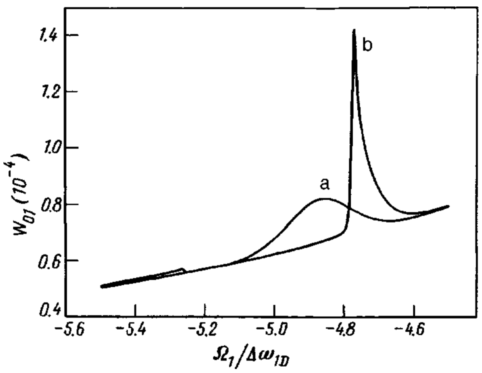

FIG. 3. Probe-field absorption resonances in the presence of another weak counterpropagating wave with different frequency ω_2: Curve a — Doppler-broadened two-photon resonance in the absence of the field E_3, b — compensation of Doppler broadening by means of a strong field E_3. The curves are normalized to the value of the linear absorption at the center of the Doppler-broadened single-photon resonance.

If the intensity of the strong field E_2 is less than optimal ($G_{12}=1.16$ GHz), then the condition for obtaining a Doppler-free resonance is not satisfied, and the height of the peak decreases while the width increases (Fig. 2, curve b). Switching on an additional strong field E_3 propagating in a direction opposite to the shortest-wavelength radiation E_2 makes it possible to reduce once again the width of the resonance to a minimum (Eq. (9)) and to increase the absorption cross section for ω_1 photons (Fig. 2, curve c). The required values are $G_{23}=2.15$ GHz and $\Omega_3=-5.04$ GHz.

5. Let us examine one more variant of sub-Doppler spectroscopy, where the intensity of the counterpropagating waves E_1 and E_2 is low, while their frequencies are chosen to be different so as to approach an intermediate resonance with level 1. In this case, the absorption cross section grows approximately by 6 orders of magnitude, but the condition for compensation of Doppler broadening at the transition 02 is not satisfied, and the two-photon absorption linewidth cannot be narrower than $|k_1-k_2|\overline{v}$ ($\overline{v}$ is the thermal velocity). When an additional strong field of frequency ω_3, propagating in the opposite direction to the wave E_2, is switched on, it becomes possible (Eq. (8)) to obtain a narrowed line having a linewidth comparable to the minimum possible two-photon absorption linewidth in weak fields (i.e., when $\mathbf{k}_1=-\mathbf{k}_2$) (Fig. 3). The illustrations correspond to the same transitions in Li with $G_{12}=8.35$ MHz and $\Omega_2=6.68$ GHz ($(k_2-k_1)/k_1\approx 0.1$). Curve a corresponds to $E_3=0$; curve b corresponds to the same conditions but in the presence of a strong field E_3 with $G_{23}=0.45$ GHz and $\Omega_3=0.94$ GHz, propagating in the same direction as the wave E_1.

6. In conclusion, we note that the possibilities examined above can be easily extended to transition schemes of the Raman-scattering type as well as to schemes where all three fields interact with a common intermediate level.

When the difference of the frequencies of the fields interacting with adjacent transitions is large, compensation of the Doppler broadening requires high intensities of the additional radiation. As the intensity of the radiation increases, the Doppler-free resonance undergoes field-induced broadening. However, this conflict can be overcome, since the magnitude of the field-induced broadening is proportional to the product of the intensities of the interacting radiation, while elimination of Doppler broadening can be accomplished by increasing the intensity of only one of the fields.

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[a]e-mail: popov@cc.krascience.rssi.ru

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