Nonlinear-resonance line shapes: Dependence on the transverse intensity distribution of a light beam

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We analyze the line shape and width of atomic coherent-population-trapping (CPT) resonances excited by laser beams with different transverse intensity profiles. A dramatic difference in the resonance line shape is found when comparing a beam with a ''steplike'' profile to a beam with a Gaussian profile. In particular, for nonuniform profiles, a non-Lorentzian functional form is given that is more appropriate for describing the nonlinear resonance line shape than is a conventional Lorentzian. Our analysis is supported by measurements of CPT line shapes in a thermal vapor of ^{85}Rb.

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The nonlinear response of a medium to resonant incident light has found a large number of applications in science and technology. Some common examples are nonlinear high-resolution laser spectroscopy [1–5], magneto-optics and magnetometry [6,7], frequency standards, and atomic clocks [8]. In all these cases, a detailed description of the line shape of the spectroscopic signal and its dependence on various parameters is of great significance in optimizing performance.

The most fundamental resonance shape, which appears in various theoretical models describing the interaction of light with atoms, is a generalized Lorentzian:

$$L(\delta) \propto A\,(\gamma^2/(\gamma^2+\delta^2)) + B\,(\gamma\delta/(\gamma^2+\delta^2)), \tag{1}$$

where γ is an atomic parameter related to the resonance width, δ is a frequency parameter of detuning from the atomic resonance, and A,B are constants independent of (or weakly dependent on) δ. In particular, such expressions appear in the theory of one-photon (with $B=0$ or $A=0$) [1] and two-photon resonances [9].

However, if additional factors are taken into account the shape of the spectroscopic signal can differ significantly from Eq. (1). For instance, in a thermal gas of free particles, one of the most significant factors is the translational motion, which affects the resonance line shape primarily through: (a) the Doppler shift of the resonance frequency for moving atoms; or (b) transit-time effects due to the finite interaction time of the atoms with a light beam of finite extent. For example, due to the Doppler effect, the one-photon absorption resonance can be significantly broadened and its shape acquires the well-known Gaussian dependence on detuning (within one Doppler width) [10]:

$$D(\delta) \propto \exp[-\delta^2/(k\overline{v})^2], \tag{2}$$

where $k\overline{v}$ is the usual parameter describing the Doppler width, in which k is the magnitude of the wave vector and $\overline{v}/\sqrt{2}$ is the root-mean-square velocity of the atoms.

This paper studies the influence of another factor on the spectroscopic signal: that of the transverse distribution of the light-beam intensity in the power-broadened regime (as opposed to the transit-time regime). We find that this important factor leads to significant changes in the resonance shape and width. Here, we compare the line shapes for two light-beam profiles with cylindrical symmetry, one steplike with a uniform intensity over a limited spatial range, and one Gaussian.

We consider this problem in the limiting case where the rate $\Gamma_{\mathcal{L}}$ of redistribution of atoms among internal degrees of freedom significantly exceeds the inverse of the average drift time through the beam in the transverse direction. Thus, if the beam has a radius r_0, the condition considered here can be written as

$$\overline{v}/r_0 \ll \Gamma_{\mathcal{L}}, \tag{3}$$

and in the presence of a buffer gas (D is the diffusion constant)

$$D/r_0^2 \ll \Gamma_{\mathcal{L}}. \tag{4}$$

The conditions (3) and (4) essentially define the ranges of parameters where the resonance width is no longer influenced by the motion of the atoms, but instead is governed primarily by the light intensity and relaxation constants in a chosen specific atomic model. In this case, moving atoms see the local field intensity change adiabatically, and the effects of the transverse motion can be ignored. If the spectroscopic signal is the absorption (or scattering) of the field propagating through the resonant medium, then we must calculate the volume integral of the excited-state population n_e. For optically thin medium and collimated light beams it is sufficient to integrate over only the transverse coordinates x and y:

$$\text{Signal} \propto \int\int n_e(\mathbf{r}_\perp)\, d\mathbf{r}_\perp^2. \tag{5}$$

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In particular, we will analyze the shape and width of nonlinear resonances, considering two light beams with the same power P, radius r_0, and cylindrically symmetric profiles of the intensity distribution (steplike or Gaussian). In this case the dependence of the intensity on the transverse distance $r=\sqrt{x^2+y^2}$ to the beam center can be written as

$$I(r)=I_0 f(r/r_0), \tag{6}$$

where the peak intensity $I_0=P/\pi r_0^2$ is the same for both beams, and $f(r/r_0)$ is a profile function $(0\leqslant f(r/r_0)\leqslant 1)$. Thus, for the steplike profile we have

$$f(r/r_0)=\begin{cases}1, & r\leqslant r_0\\ 0, & r>r_0,\end{cases} \tag{7}$$

and for the Gaussian profile we have

$$f(r/r_0)=\exp\{-(r/r_0)^2\}. \tag{8}$$

Even without an exact analysis we can see that the resonance width in the case of a Gaussian profile must be lower than in the case of a steplike profile, when intensity effects are taken into account. For the Gaussian beam, the intensity reaches its peak value, I_0, only at the very center $(r=0)$ of the beam; while for the steplike profile this peak intensity is present everywhere inside the beam. If we remember that we are in the limit of atoms more or less ''stuck'' in the buffer gas, this means that all atoms in the steplike profile see the same intensity, whereas for the Gaussian beam, the change in line shape is inhomogeneous in nature, and atoms in the outer regions contribute narrower lines to the overall shape. This simple observation clearly demonstrates why the profile function is important when analyzing experimental dependencies. However, in practice the field intensity is usually calculated as P/s (s is the area of the light spot), without a detailed investigation of the intensity profile. Thus, for seemingly identical parameters, different experimental setups can produce significantly different results.

Let us consider an important example connected with two-photon Raman spectroscopy, where a coherent interaction between two monochromatic fields takes place, such as in the three-level Λ system. It is well known that the coherent population trapping (CPT) effect can occur in this scheme [11]. The CPT leads to the appearance of narrow spectral features as a function of the frequency difference between the two lasers. Such resonances are usually called dark resonances, or CPT resonances. For example, from the solution of the steady-state equation for the density matrix of a Λ atom (see Fig. 1) it follows that in the low-saturation limit, $V_j^2\ll 1$, with respect to one-photon transitions and for small one-photon detuning, $\delta_L\ll\gamma$, the two-photon contribution to the excited-state population $\tilde{n}_e$ can be written as

$$n_e^{2\mathrm{ph}}\propto\frac{V_1^2V_2^2[\tilde{\Gamma}+2(V_1^2+V_2^2)]}{[\tilde{\Gamma}+2(V_1^2+V_2^2)]^2+\tilde{\delta}_R^2}. \tag{9}$$

Here $V_j=dE_j/\hbar\gamma$ is the normalized Rabi frequency of the jth wave $(j=1,2)$, $\tilde{\Gamma}=\Gamma/\gamma$ is the normalized relaxation rate of the ground-state coherence (for example, due to collisions with a buffer gas), $\tilde{\delta}_R=\delta_R/\gamma$ is the normalized two-photon detuning, and γ is the homogeneous width of the optical transition. Further approximations in Eq. (9) are $\tilde{\Gamma}\ll 1$ and $\tilde{\delta}_R\ll 1$, which are very well fulfilled in experiments. Equal probabilities (1/2) are also assumed for the spontaneous transitions from the excited state $|e\rangle$ to each of the lower states $|1\rangle$ and $|2\rangle$.

Most conveniently, the bichromatic field for the two-photon spectroscopy in the Λ scheme is provided by modulating the frequency of a single laser. Thus we can assume the same profiles for both waves. The analysis of the resonance shape and its dependence on the total resonant power (for a cylindrically symmetrical profile) then begins with the expression

$$\mathcal{R}=2\pi r_0^2\int_0^\infty\frac{wf^2(w)V_1^2(I_0)V_2^2(I_0)[\tilde{\Gamma}+2(V_1^2(I_0)+V_2^2(I_0))f(w)]}{[\tilde{\Gamma}+2(V_1^2(I_0)+V_2^2(I_0))f(w)]^2+\tilde{\delta}_R^2}dw \tag{10}$$

for the line shapes, where $V_j^2(I_0)$ corresponds to the peak light intensity. Assuming that the intensity ratio is constant $[V_1^2(I_0)/V_2^2(I_0)$ does not depend on $I_0]$ we arrive at the expression

$$\mathcal{R}=2\pi r_0^2\int_0^\infty\frac{wf^2(w)S_0^2[1+S_0f(w)]}{[1+S_0f(w)]^2+\Delta^2}dw. \tag{11}$$

Here $S_0=2[V_1^2(I_0)+V_2^2(I_0)]/\tilde{\Gamma}$ is proportional to the peak intensity I_0 and $\Delta=\tilde{\delta}_R/\tilde{\Gamma}=\delta_R/\Gamma$. Obviously, the dependence of the resonance on light intensity is governed by the dependence of Eq. (11) on S_0.

The integral Eq. (11) is easily found for the steplike profile of the light beam:

$$\mathcal{R}_{\mathrm{st}}=\pi r_0^2\{S_0^2[1+S_0]\}/\{[1+S_0]^2+\Delta^2\}, \tag{12}$$

and represents the usual Lorentzian curve as a function of normalized detuning Δ. For the Gaussian profile another expression results:

$$\mathcal{R}_G=\pi r_0^2(S_0-\Delta\arctan[S_0\Delta/(1+S_0+\Delta^2)]+\tfrac{1}{2}\ln\{(1+\Delta^2)/[(1+S_0)^2+\Delta^2]\}). \tag{13}$$

As seen from Fig. 2(a), the resonance shape becomes sharper for the Gaussian profile compared to the steplike intensity distribution. The explanation here is: the central part of the line is due to the contribution of atoms in the spatial region where the light intensity is small and where the resonance

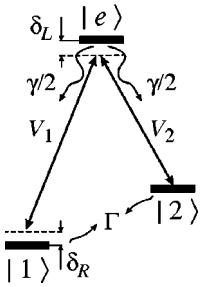

FIG. 1. Three-level Λ system with the nomenclature used in this paper.

width corresponds to the collisional width Γ. The resonance curvature in the center of the absorption curve as a function of δ is governed mainly by this width Γ. The dependence of the resonance amplitude on intensity [Fig. 2(b)] is very similar for the two cases, and for large S_0, the slopes of the two lines are equal ($\propto S_0$); the only difference being a constant offset. It is also seen that for the Gaussian profile the transition to the region of linear dependence is smoother and that it occurs at larger intensity I_0.

There is a significant difference in magnitude of the resonance width, as seen in Fig. 2(c). Furthermore, for the Gaussian profile the dependence is nonlinear (this is hard to see from viewing the figure). Though in the general case a simple analytical expression for the width of the resonance described by Eq. (13) does not exist, for large intensity we can nevertheless use the simplified asymptotic expression for the line shape:

$$\mathcal{R}_G \propto \{1-(\Delta/S_0)\arctan(S_0/\Delta)\}, \quad S_0 \gg 1. \tag{14}$$

From this we find that at large field intensity the resonance width

$$(\mathrm{FWHM})_G/\Gamma \approx 0.86 S_0, \tag{15}$$

which is less than half that for the beam with steplike profile, where

$$(\mathrm{FWHM})_{\mathrm{st}}/\Gamma \approx 2 S_0, \tag{16}$$

although the resonance amplitude is practically the same in both cases. In Fig. 3 the new dependence seen in Eq. (14) is evident. It has a sharp tip in the center and an asymptotic behavior $\sim 1/\Delta^2$ at large detuning.

Equations (13) and (14) with S_0 as a fitting parameter are useful for numerical data processing in experiments with laser beams of Gaussian profile, such as two-photon spectroscopy or various magneto-optical resonances in a monochromatic light field (e.g., Hanle spectroscopy, nonlinear Faraday effect), where the Zeeman splitting of the ground state takes the role of the two-photon detuning δ_R.

The theoretical treatment of the preceding sections was verified in part by measurements of CPT line shapes in a thermal Rb vapor confined in a cell with a buffer gas. The experimental setup is shown in Fig. 4. A distributed-Bragg-reflector diode laser tuned to the D_1 transition in ^{85}Rb at 795 nm was modulated with a rf signal at 1.5 GHz. This modulation produced sidebands (of unequal intensity) on the optical carrier separated from each other by 3.0 GHz, roughly the ground-state hyperfine splitting of ^{85}Rb. At the appropriate

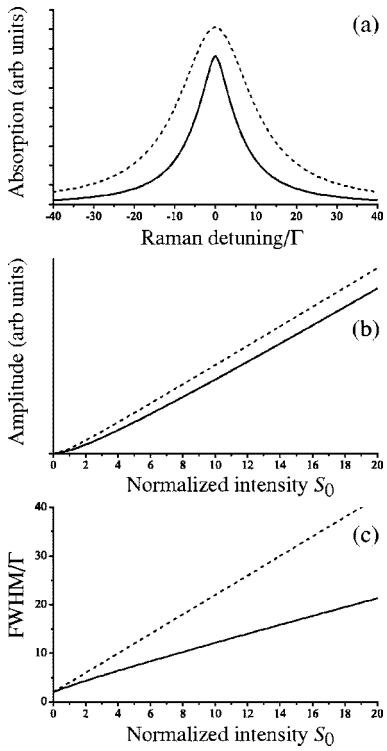

FIG. 2. The two-photon contribution in the excited-state population of three-level atoms under bichromatic excitation. Comparison of the Gaussian (solid curves) and steplike (dashed curves) beam profiles. (a) The line shapes at $S_0=10$. (b) The resonance amplitude versus intensity. (c) Full width at half maximum (FWHM) versus intensity.

optical tuning, a Λ system was formed by the two first-order optical sidebands, connecting the $F=2$ and $F=3$ hyperfine levels of the $5S_{1/2}$ ground state to the $5P_{1/2}$ excited state. Light from the diode laser was collimated and sent through a quarter-wave plate to circularly polarize the light. The beam then entered a glass cell containing a thermal vapor of Rb with the natural abundance of the two isotopes, and a buffer gas of 0.4 kPa of Ne. At this buffer gas pressure, the Rb atoms are in a predominantly diffusive regime, described well by Eq. (4). The cell was surrounded by a magnetic shield, and a weak longitudinal magnetic field was applied with a pair of Helmholtz coils. Finally, the power of the light transmitted through the cell was detected by a photodiode (PD) to measure the CPT resonance. The optical absorption

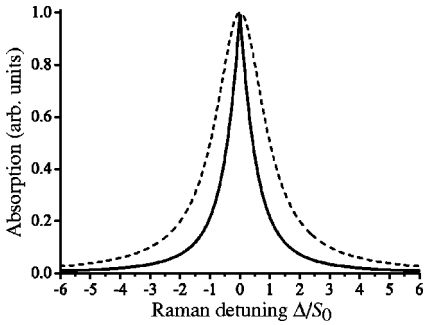

FIG. 3. Comparison of the asymptotic line shape for the Gaussian beam profile Eq. (14) (solid curve) and the usual Lorentzian shape $1/(1+(\Delta/S_0)^2)$ (dashed curve).

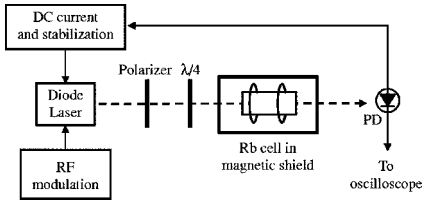

FIG. 4. Experimental setup for the measurement of the CPT line shape using a modulated laser diode

at the peak of the Doppler-broadened optical resonance was 12%, indicating that the cell was optically thin.

The spatial profile of the light was measured near the cell position. The vertical profile showed a close match to a step function (probably because of clipping of the light beam by the lens used to collimate the laser light), and the horizontal profile showed an even closer match to a Gaussian profile. Thus the situation closely resembles the intensity profile given by Eq. (7), at least in one dimension. In order to study the asymptotic behavior an average intensity of 160 μW/cm^2 was used in the experiments, much larger than is typically used in precision experiments with CPT resonances [12–14].

When the frequency of the laser current modulation was scanned near the first subharmonic of the atomic ground-state hyperfine transition, a change in the absorption of the light in the vapor cell was observed, typical of a CPT resonance. A typical resonance is shown in Fig. 5. Also shown are fits to the model line shapes for the two types of spatial profiles discussed earlier, with the width and amplitude as fitting parameters. It can be seen from the residuals that the fit to the arctan line shape of Eq. (14) is clearly superior to that of the simple Lorentzian. The experimental data therefore support the theoretical treatment provided earlier.

We have described theoretically how the line shape of CPT resonances in a Λ system depends on whether the transverse profile of the excitation light is a step function or a Gaussian. For a step-function profile, the usual Lorentzian line shape results, while for a Gaussian profile the line shape is described by an expression involving an arctan function. In addition, the Gaussian beam profile results in a narrower transition width, which could lead to improved sensitivity or stability for magnetometers or atomic frequency references based on CPT, although in many cases the best strategy is to fill the entire cell uniformly with the beam. However, the relevance of this work to the design of such devices is enhanced by the fact that lasers used to measure the atomic resonances often naturally have Gaussian beam profiles. As a result, frequency references or magnetometers using laser-driven CPT resonances can be easily designed to use Gaussian beam profiles. In addition, under some experimental conditions, the sharply peaked structure of the arctan line shape for Gaussian beam excitation may enable a more precise determination of the resonance center.

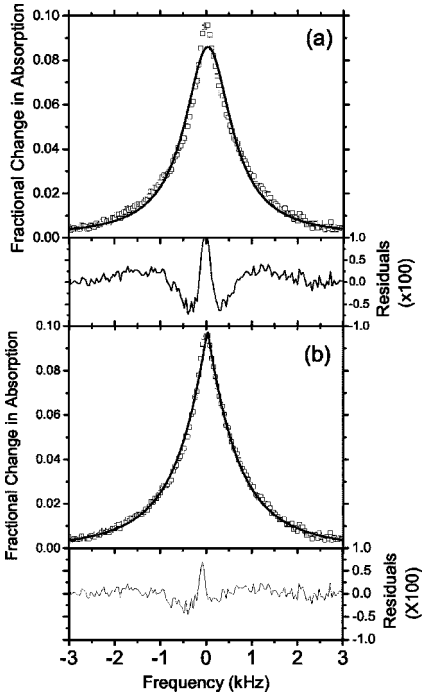

FIG. 5. Line shape measured using a laser beam with a Gaussian spatial profile compared with theoretical predictions. The open squares are the experimental data points. (a) shows a best fit to a simple Lorentzian line shape, while (b) shows a best fit to the arctan profile of Eq. (14). The residuals from the fit are shown below each plot. The width and height are independent parameters in the fitting process.

Experimental measurements are also presented on CPT resonances in a thermal vapor of ^{85}Rb excited by a beam with a Gaussian profile in one transverse dimension. For these measurements, Eq. (14) provides a better fit to the experimental line shapes than does a simple Lorentzian. The experimental data therefore supports the theoretical results for excitation with Gaussian beams.

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