How weak is a weak probe in laser spectroscopy?

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Laser spectroscopy experiments are often conducted with a probe that does not significantly alter the medium's properties. For a two-level atom a clear measure of the strength of a probe beam in terms of the saturation intensity is known. We show that for a multilevel atom the situation is very different, and the effects of optical pumping are crucial to understanding the opacity of the medium. We present a simple theoretical analysis for Doppler-broadened spectroscopy of alkali metals on the D_2 line that emphasizes the importance of the transient nature of the population dynamics, and the crucial role of the distribution of the times of flight of atoms through the probe beam. Experimental results are obtained with room temperature rubidium vapor probed by an extended-cavity diode laser and confirm our theoretical prediction. © *2009 American Association of Physics Teachers.*
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I. INTRODUCTION

Laser spectroscopy is a powerful diagnostic tool. The intensity, shape, and frequency of the spectral features yield valuable information about a medium, including macroscopic parameters such as pressure and temperature to microscopic population distributions among internal states of atoms and molecules. It is often possible to operate in a regime where probing by the laser yields valuable information, without the medium being significantly perturbed by the light.

One of the simplest systems to study by Doppler-broadened spectroscopy is a metal vapor realized with a single monochromatic probe beam. From an experimental perspective rubidium is an ideal atom to study, with sufficient vapor pressure at room temperature and a strong resonance line that can be accessed with inexpensive and reliable laser diode sources. Consequently, Rb has been used extensively for pedagogical experiments to demonstrate topics in linear and nonlinear laser spectroscopy.[1–10] The absorption spectra of room temperature gases are among the easiest to analyze theoretically—the thermal motion of atoms in the vapor produces a range of frequencies (Doppler broadening) which exceeds the natural linewidth of the atomic transition, and the density is typically so low that collisions between atoms can be ignored. The system examined in this paper is the Doppler-broadened absorption spectroscopy of Rb on the D_2 line $5^2S_{1/2} \rightarrow 5^2P_{3/2}$.

Although absorption spectroscopy of a two-level atom is discussed in textbooks, for example, Ref. 11, we will demonstrate that this model is insufficient to explain simple Doppler-broadened spectra of multi-level atoms. We will discuss the underlying mechanisms that govern the form of probe absorption spectra for multi-level atoms. In particular, we show that even a simple question such as "how weak must a weak-probe beam be?" has a different answer than the one most frequently encountered. In the pedagogical experiments described in Refs. 1–4, 8, and 10 the effect being studied is typically largest when the probe beam is weak. In Refs. 2, 4, 8, and 10 explicit comparisons are made between theory and experiment for the size and shape of absorption features, and it is shown that the agreement is significantly better if the experiments are realized in the weak-probe limit.

The paper is organized as follows. Section II presents the theoretical model, evolving from a two-level atom to a multilevel atom, and discusses the incorporation of the longitudinal and transverse motion of the atoms. In Sec. III the experimental method is presented, and results are shown and discussed in Sec. IV. Concluding remarks are made in Sec. V.

II. THEORETICAL MODEL

A. Theory for stationary two-level atoms

We assume a monochromatic laser of angular frequency ω_L interacting with a medium composed of two-level atoms. The energy separation between the ground and excited states is $\hbar\omega_0$, and the detuning parameter Δ is defined as $\Delta = \omega_L - \omega_0$. The excited state b decays spontaneously to the ground state a with a rate Γ, and the corresponding lifetime $\tau = 1/\Gamma$.

Following the notation of Ref. 12 we give the following equation for the populations N_a and N_b of states a and b, respectively, in terms of the light intensity I and the saturation intensity I_{sat}:

$$\frac{dN_b}{dt} = \frac{\Gamma}{2}\frac{I}{I_{sat}}\frac{(N_a - N_b)}{1 + 4\Delta^2/\Gamma^2} - \Gamma N_b. \tag{1}$$

The terms on the right-hand side of Eq. (1) have the following physical interpretation: the excited state population grows by absorption and is proportional to the light intensity and the ground state population. Stimulated emission out of the excited state, which is also proportional to the light intensity, reduces the excited state population. The last term is spontaneous emission out of the excited state. The saturation intensity, I_{sat}, is[13]

$$I_{sat} = \frac{\hbar\omega_0}{2\sigma_0\tau}, \tag{2}$$

where the resonant excitation cross-section is given by

$$\sigma_0 = \frac{3\lambda^2}{2\pi}, \tag{3}$$

with λ the wavelength of the transition.

The crucial parameter for a two-level atom is the saturation intensity. The latter has the physical interpretation that a beam is weak if less than one photon per atomic cross-

section per excited state lifetime is incident. For a multilevel system other time scales are introduced that change the effective saturation intensity.

Because the total number of atoms $N_{\text{tot}}=N_a+N_b$ is fixed, it is easy to solve for the time dependence of the populations. The ratio of the number of atoms in the excited and ground state in equilibrium as a function of the detuning and the dimensionless parameter I/I_{sat} is (see, for example, Ref. 11, Sec. 13.3)

$$\frac{N_b}{N_a}=\frac{1}{2}\frac{\frac{I}{I_{\text{sat}}}}{1+\frac{I}{2I_{\text{sat}}}+4\frac{\Delta^2}{\Gamma^2}}. \tag{4}$$

The solution shows that if there is no incident light, the atoms reside in the ground state. As the light intensity increases, the fraction of atoms in the excited state initially grows linearly with intensity; when the light is very bright the populations saturate to a value of $1/2$. The saturation intensity is defined as the value at which the population is half of its asymptotic value for resonant excitation. The definition of "weak" in this context is clear—it is the requirement that $I \ll I_{\text{sat}}$.

The macroscopic properties of the medium can be related to the microscopic populations.[14] Specifically, the absorption coefficient, α, is given by $\alpha=(n_a-n_b)\sigma_0$, with n_a and n_b the number densities of the atoms in the ground and excited states, respectively. For the case of weak light, $N_a \sim N_{\text{tot}}$ and $N_b \sim 0$, and α can be taken as a constant for each frequency. This assumption leads to the well-known Beer-Lambert law[14] for exponential absorption as a function of distance z in the medium: $I=I_0 \exp(-\alpha z)$. When the light is intense, the approximation that all the atoms are in the ground state breaks down, and the familiar exponential decay is replaced by a polynomial dependence on distance. However, the focus of our attention will be on the weak-field limit.

B. Including longitudinal atomic velocity

The experimental spectra are Doppler broadened due to the range of longitudinal velocities. It is easy to incorporate this effect into the theoretical framework. For an atom moving along the direction of propagation of the laser with velocity component v we replace the detuning parameter Δ by $(\Delta-kv)$, where k is the wavevector of the light. The absorption profile of the medium is then obtained by summing over all possible velocities. This averaging process leads to a Voigt profile.[14] In our case the Doppler broadening is two orders of magnitude wider than the natural broadening, and therefore the absorption follows a Gaussian profile.

The line-center behavior of Doppler-broadened saturated absorption is treated in many textbooks, see, for example, Ref. 11, and is summarized here. The effect of the velocity averaging is to increase the saturation intensity, because fewer atoms interact with the probe beam. The Lorentzian line-center absorption becomes $1/(1+I/I_{\text{sat}})$, and the power broadened width grows as $\sqrt{1+I/I_{\text{sat}}}$. Therefore the Doppler-broadened line-center absorption decreases as $1/\sqrt{1+I/I_{\text{sat}}}$. The different behavior of the Lorentzian and Gaussian line-center absorption is shown in Fig. 1. Note that the absolute value of the line center absorption is also different because fewer atoms are close to resonance for the Doppler broadened case. An estimate for the reduction of the line center absorption is the ratio of the natural width, Γ, to the Doppler-broadened width. This reduction is approximately two orders of magnitude for Rb on the D lines at room temperature.

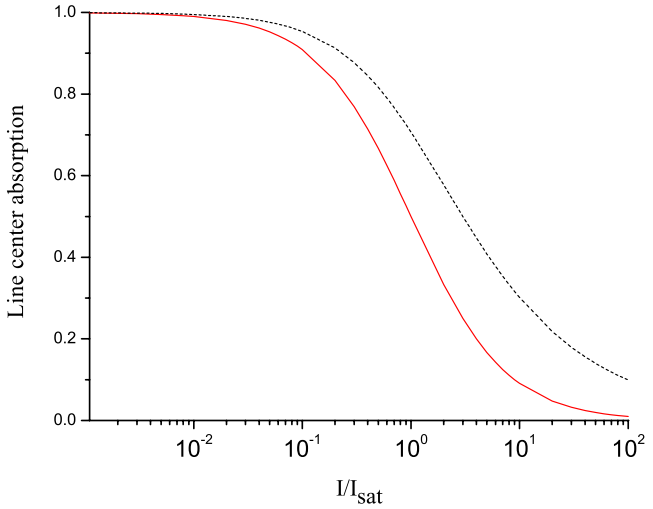

Fig. 1. (Color online) Saturation characteristics of a Lorentzian (solid line) æand Gaussian (black dashed) absorption line.

C. Extension to other levels

The most significant deviations from the model in Sec. II arise when another ground state is introduced. The third state, c, is assumed to differ significantly in energy from a (the energy splitting exceeds the Doppler width) such that light resonant with the a to b transition cannot be absorbed by atoms in state c. (State c is often referred to as a "dark state.") The new process which alters the dynamics of the system is the hyperfine or optical pumping. That is, atoms excited from a to b by the probe beam can spontaneously emit into state c, from which they will not return.

A rate-equation approach to the three-level model yields insight into the system's evolution; see, for example, Refs. 15 and 16. To gain an appreciation of the importance of hyperfine pumping we present a simple toy model of a medium of stationary three-level atoms, where the $a \rightarrow b$ transition is driven resonantly, and state b can either decay back to a at a rate $R_{ba}\Gamma$, be stimulated back to a, or spontaneously fall into state c at a rate $R_{bc}\Gamma$. A more realistic model would include all the excited states, but that is beyond the scope of this work.

The rate equations for the population changes are

$$\frac{dN_a}{dt}=R_{ba}\frac{\Gamma}{2}\frac{I}{I_{\text{sat}}}\frac{(N_b-N_a)}{1+4\Delta^2/\Gamma^2}+R_{ba}\Gamma N_b, \tag{5}$$

$$\frac{dN_b}{dt}=R_{ba}\frac{\Gamma}{2}\frac{I}{I_{\text{sat}}}\frac{(N_a-N_b)}{1+4\Delta^2/\Gamma^2}-\Gamma N_b, \tag{6}$$

$$\frac{dN_c}{dt}=R_{bc}\Gamma N_b. \tag{7}$$

Figure 2 shows the evolution of the population of the three states over a time of 25 μs, which is approximately three orders of magnitude longer than the excited state lifetime. The branching ratios R_{ba} and R_{bc} were chosen to have the

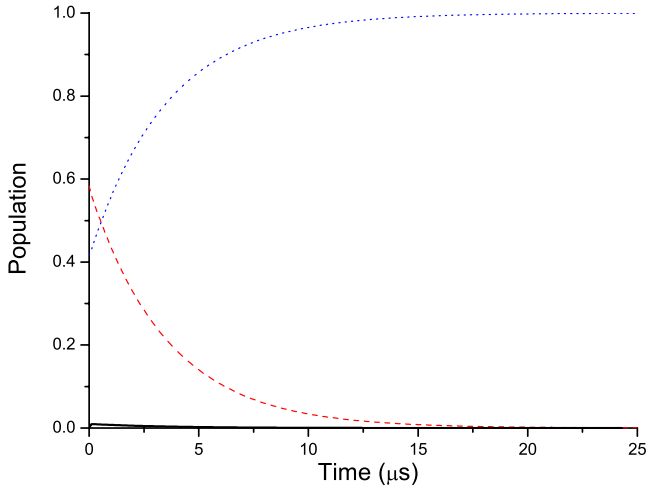

Fig. 2. (Color online) Population dynamics for a three-level atom for $I/I_{sat}=0.1$. There is optical pumping of the population from state a (dashed) to state c (dotted), although the excited state population (solid line) is always small.

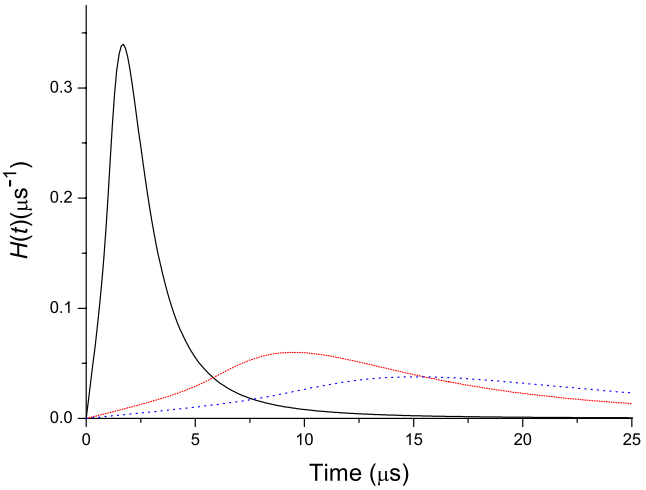

Fig. 3. (Color online) Probability distribution of the time of flight for a beam width of 0.3 mm (solid line), 1.7 mm (dotted), and 2.7 mm (dashed).

values for the ^{85}Rb $F=3\rightarrow F'=3$ and $F=2\rightarrow F'=3$ transitions, respectively, because the experiments were conducted with this transition. The initial conditions reflect the degeneracies of the states. There are seven sublevels for the $F=3$ state a and five sublevels for the $F=2$ state c. The solutions are shown for $I/I_{sat}=0.1$.

There are two important differences for the dynamics when decay to a third level is incorporated. First, for weak probe beams the time scale of the optical pumping process becomes very long. Second, although the population of state b is always small, there can be a transfer of population from a to c. Consequently, there will be no absorption of radiation resonant with the a to b transition when the system achieves equilibrium; it is the transient behavior of the dynamics which governs the absorption.

D. Including the transverse atomic velocity

The manifestation of transverse atomic motion is the finite time of flight for an atom through the (finite width) probe beam. By considering the distribution of path lengths it is possible to derive a closed form for $H(t)$, the probability distribution for an atom traversing the beam with a time-of-flight t; see, for example, Eq. (11) of Ref. 12, or Eq. (8) of Ref. 17.

Figure 3 shows the dependence of $H(t)$ on the beam radius. The beam radii were chosen to correspond to the widths of the Gaussian beams used in the experiment. For the smallest beam used with radius 0.3 mm the most probable time of flight is 1.7 μs, rising to 9 μs for a 1.7 mm beam and 15 μs for a 2.7 mm beam. It is instructive to compare these typical times with the time scale for population dynamics from Fig. 2. Although for $I/I_{sat}=0.1$ the two-level atom theory predicts that 95% of the atoms are still in state a, we see that there is considerable evolution of the population within 25 μs. The pumping into the dark state c is significant, and the absorption of the medium will be reduced dramatically. For the narrowest beam most of the atoms spend less than 10 μs in the beam, and there is considerably less optical pumping. Figure 3 shows that for a given probe intensity, the size of the beam is likely to have a pronounced effect on the absorption of the medium, and that wider beams will experience reduced absorption as a consequence of optical pumping.

III. EXPERIMENT

We designed an experiment to test our prediction of a drastic change in the absorption with beam width. The apparatus shown in Fig. 4 consists of a grating-tuned and spectrally narrowed extended-cavity diode laser similar to the design of Arnold *et al.*[18] The laser has a Sharp GH0780MA2C chip, collimated with a 4.5 mm focal length, aspheric lens, to give an output beam with a spot radius (half width at half maximum) of $w=0.30\pm0.02$ mm vertically and (0.61 ± 0.02) mm horizontally. A pair of anamorphic prisms was used to reduce the horizontal beam width such that the beam was circular to 95%.

Neutral density filters were used to attenuate the probe power. The probe beam was focused onto a photodiode cir-

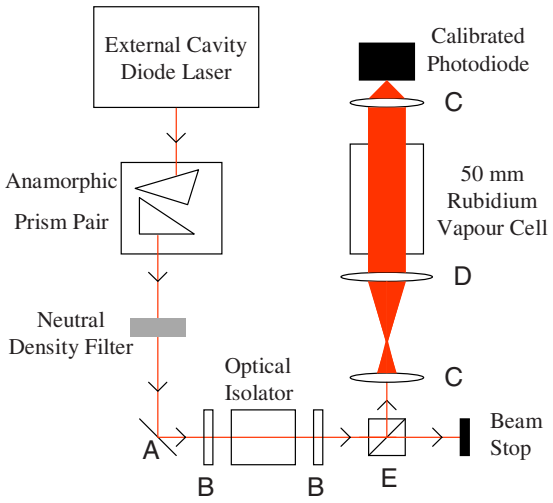

Fig. 4. (Color online) Optical layout. The combination of neutral density filters and half-wave plates B is used with the polarizing beamsplitter E to vary the power of the probe beam. The 50 mm focal length lenses C and different lenses D are used in a telescope to vary the width of the probe beam in the cell.

cuit that recorded a voltage linearly proportional to the incident intensity. Saturated absorption/hyperfine pumping spectroscopy[1,8] was used for frequency reference. Different focal length lenses were used in the telescope to change the magnification of the expanded beam. The 50 mm long vapor cell contained ^{85}Rb and ^{87}Rb in their natural abundances at room temperature. No measures were taken to remove any residual magnetic field within the vapor cell. For a Doppler-broadened D_2 absorption line the transmitted probe intensity was measured at line center far from resonance to obtain the 100% transmission level. A reading was also taken with the probe beam blocked to ascertain the effects of stray light. The line center percentage absorption was calculated from these readings.

Three different combinations of lenses were used in the telescope to yield beam widths w of (0.30 ± 0.02) mm, (1.70 ± 0.02) mm, and (2.72 ± 0.04) mm. From the measured beam power, P, and width w, the peak intensity at the entrance of the cell, I_0, is calculated from the relation [see, for example, Ref. 15. Eq. (14)]

$$I_0 = \frac{P \ln 2}{\pi w^2}. \tag{8}$$

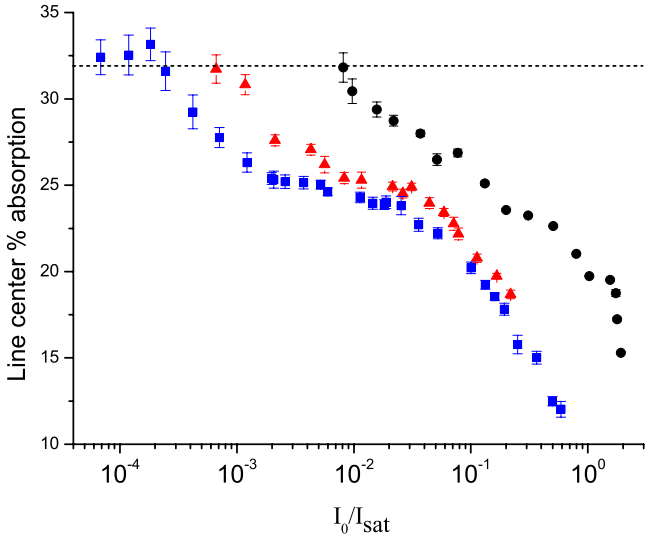

Fig. 5. (Color online) The line center absorption for the ^{85}Rb $F=3\rightarrow F'$ transition is plotted as a function of I_0/I_{sat}, with I_0 being the peak intensity at the entrance of the cell. Three beam widths were used: 0.3 mm (circles), 1.7 mm (triangles), and 2.7 mm (squares). The theoretical prediction is shown as a dashed line.

IV. RESULTS AND DISCUSSION

The D_2 line in Rb has four Doppler-broadened transitions, two per isotope. The hyperfine interaction produces two F values for the 5 $s^2S_{1/2}$ ground term, with an energy separation which exceeds the room temperature Doppler width. In contrast, for the $5p^2P_{3/2}$ excited term the hyperfine intervals of the four F' levels is smaller than the Doppler width. The absorption at the line center for all four Doppler-broadened transitions was measured as a function of probe power and intensity for different beam widths. For the remainder of this paper only data from the ^{85}Rb $F=3\rightarrow F'$ transition is shown because the other transitions show largely the same dependence.

Figure 5 plots the line center absorption as a function of I_0/I_{sat} for three beam widths (note the logarithmic scale for the beam intensity). For this transition the expected line center absorption (based on the measured vapor pressure and the measured transition dipole matrix elements) at room temperature in a 50 mm long cell in the weak-probe limit is 32%.[19] Two effects are readily apparent. For a fixed intensity but different beam width, the absorption is different. Secondly, the intensities at which the absorption agrees with the weak-probe limit are orders of magnitude lower than those expected from the Doppler broadened two-level atom model encapsulated in Fig. 1. As expected, there is a reduction of absorption for intensities $I \geqslant I_{sat}$, but the details of strong-field saturation of absorption are beyond the scope of this paper.

For a given beam intensity there is larger absorption and closer agreement with the theoretical prediction for smaller beam diameters. This behavior is consistent with the analysis of Sec. III—atoms spend less time in narrower beams, and the deleterious effects of optical pumping are less significant. Figure 5 shows that values of $I/I_{sat} \leqslant 10^{-3}$ are required for the widest beam before the weak-field limit is obtained. Note also that for the smallest beam diameter, power levels of the order of 100 nW are needed to ensure that the probe beam is weak.

Attempts have been made[15,16] to modify Eq. (2) to take into account the different time scales when optical pumping is present. The two new time scales are the characteristic time for optical pumping and the transit time through the beam. Because both are longer than the excited state lifetime, a lower saturation intensity is predicted. We do not utilize such an approach here for two reasons: there is a range of transit times and averaging over them is crucial to gain an understanding of the absorption of the medium; the usefulness of the concept of saturation intensity has to be questioned. It is not enough to classify the probe beam by a single dimensionless parameter, and the power and beam radius have to be specified. Also we need to know the temperature and atomic mass to take into account the speed of the atoms. Using the equilibrium values for populations is an excellent approximation for a two-level atom, and knowledge of I/I_{sat} alone specifies the absorption. However, adding the extra mechanism of optical pumping to a (dark) ground state completely changes the dynamics. The equilibrium situation has no absorption, hence an understanding of the transient nature of the dynamics is crucial for modeling the absorption of the medium. The specification of a weak-probe beam is thus in terms of the time scale for optical pumping compared with the ranges of the time of flight of atoms through the medium and cannot be specified by a single dimensionless parameter.

The reduction of absorption due to optical pumping should be absent for a closed transition, that is, one where the selection rules prevent atoms from falling into the dark state. In the transition considered here, that would correspond to $F=3\rightarrow F'=4$. However, as was noted in Ref. 8 there is significant optical pumping due to the presence of the open $F=3\rightarrow F'=3,2$ transitions and the hyperfine splitting of the excited states being less than the Doppler width. For the D_1 line $(n^2S_{1/2}\rightarrow n^2P_{1/2})$ there is no closed transition, and the effects of optical pumping are likely to be more pronounced.[15]

In this work we concentrated on analyzing the line-center absorption. Each Doppler broadened line is a composite of three transitions due to the $\Delta F=0, \pm 1$ selection rule. Optical

pumping will influence these lines to a different extent. Therefore, the shape of the Doppler broadened transitions also evolves as the probe beam is made weaker.

V. CONCLUDING REMARKS

There have been previous studies of the effect of the time of flight in nonlinear spectroscopy.[20–22] Our aim in this paper has been to highlight that great care is needed to interpret Doppler-broadened absorption spectra where there are pumping processes with time scales comparable to the distribution of times of flight. In most pump-probe experiments the central idea is to investigate the difference in propagation of the probe pulse through a medium due to the presence of a pump beam. The best signal to noise ratio is usually obtained with the largest probe-only absorption. To distinguish clearly the extra effects of the pump beam the probe beam must be sufficiently weak so that it does not self-pump the medium. The absence of pumping by the probe is particularly important in experiments where the redistribution of the population among the ground state sublevels is the signature of the effect being studied, for example, polarization spectroscopy[23] and saturated-absorption spectroscopy.[24]

In conclusion, we have presented a simple theoretical argument which shows that for a multi-level atom the time scale for optical pumping can be much longer than the typical time of flight of an atom through a probe beam. Although there is negligible population in the excited state, significant transfer of population into another (dark) ground state occurs. The concept of saturation intensity was found to have limited utility under these conditions, and the definition of a weak probe beam was shown to depend crucially on the atomic velocity distribution and the probe beam size. These predictions were verified by experiment.

ACKNOWLEDGMENTS

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