Excitation dependence of resonance line self-broadening at different atomic densities

Hebin Li\(^1\), Vladimir A Sautenkov\(^{1,2}\), Yuri V Rostovtsev\(^1\) and Marlan O Scully\(^{1,3}\)

\(^1\) Department of Physics and Institute for Quantum Studies, Texas A&M University, College Station, Texas 77843-4242, USA
\(^2\) P.N. Lebedev Institute of Physics, Moscow 119991, Russia
\(^3\) Applied Physics and Materials Science Group, Engineering Quad, Princeton University, Princeton, NJ 08544, USA

E-mail: lihebin@tamu.edu

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Abstract

We study the dipole–dipole spectral broadening of a resonance line at high atomic densities when the self-broadening dominates. The selective reflection spectrum of a weak probe beam from the interface of the cell window and rubidium vapour is recorded in the presence of a far-detuned pump beam. The excitation due to the pump reduces the self-broadening. We found that the self-broadening reduction dependence on the pump power is atomic density independent. These results provide experimental evidence for the disordered exciton based theory of self-broadening, and can be useful for the description of the interaction of a strong optical field with a dense resonance medium.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

For many applications and fundamental physics it is necessary to know the nonlinear optical response of a resonance atomic gas under conditions when dipole–dipole interactions between atoms in the ground and excited states cannot be neglected.

There was a common opinion that calculations of self-broadening can be performed by using the two-particle approximation in the fast collision limit (impact collisions) as well as in the opposite limit, static interactions [1–4]. In [4], the theory of self-broadening is developed on the basis of disordered exciton in a dense resonance medium, in which many particle interaction should be taken into account. By using this model it was shown that the self-broadening is a combination of collision and static atomic interactions. A ratio of the static width to the collision width is independent of atomic density in a wide range where thermal motion of atoms can be neglected.

It has been shown that the probing of the homogeneous and inhomogeneous contributions to the linewidth can be performed efficiently by nonlinear optical methods such as photon echoes and hole burning [5, 6]. The inhomogeneous component of the spectral line could be sensitive to the optical saturation. Recently by using time resolved femto-second spectroscopy, the non-Markovian collision dynamics and the bi-exponential correlation of energy level fluctuations has been observed in a dense potassium vapour and simulations of molecular dynamics are in good agreement with experimental results [7]. The slow exponential component is attributed to long-range resonant attraction in a dense atomic vapour. By using the CW pump-probe technique, the excitation dependence of the self-broadening is observed in rubidium [8, 9] and potassium [10, 11] vapours. Nevertheless, note that in these papers the measurements have been performed only at selected atomic densities.

In the current paper the selective reflection spectrum from the interface between the cell window and rubidium vapour are recorded in the presence of a far-detuned pump beam. We have studied the excitation dependence of self-broadening of a resonance atomic line at different atomic densities in the range...
where self-broadening of atomic line is stronger than Doppler broadening. The excitation dependence of self-broadening is found to be independent of the atomic density. These results support the disorder exciton based theory of self-broadening [4].

2. Experiment

The experiment was performed with a pump-probe scheme shown in figure 1. The reflectivity and frequency-modulated (FM) reflectivity spectra of a dielectric–vapour (rubidium) interface were measured at the $5^2S_{1/2} \rightarrow 5^2P_{3/2}$ transition ($D_2$ line, shown in figure 2) of rubidium atoms. Please note that the excited state hyperfine structure is not resolved in our experiment since the splittings are less than the Doppler width (0.5 GHz) at room temperature. The probe laser is a free running diode laser (linewidth 20 MHz) which can be scanned over 30 GHz around $D_2$ transition. The frequency of the probe laser is calibrated by reference to the absorption of a rubidium cell at room temperature. The power of the probe beam is small enough (less than 100 $\mu$W) such that no saturation effects need to be considered. The pump laser (an extended cavity diode laser) provides a laser beam with power up to 180 mW, and it is far-tuned (20 GHz) to the red wing of $D_2$ line to avoid coherent effects. The frequency of the pump laser is determined by beating with the probe laser. Both pump and probe beams are focused down to a spot with 100 $\mu$m diameter by a 15 cm lens, and they are overlapped at the inner surface of the cell window.

The cell contains natural abundance of rubidium vapour. The cell is made of sapphire, and the windows are Garnet crystal which is free of birefringence. The cell can be heated to reach a high atomic density ($N \approx 10^{17}$ cm$^{-3}$). The windows are slightly wedged in order to separate the reflections from two surfaces.

The reflected beam from the interface between the rubidium vapour and the window is sent to a photodetector (PD). The signal from the photodetector is processed by a lock-in amplifier while we frequency modulate the probe laser with modulation depth of 37 MHz at frequency of 8 kHz. FM reflectivity spectra were used to improve the signal-to-noise ratio in our experiment, and it can reveal subtle details of change in reflectivity. A typical FM reflectivity spectrum is shown in figure 3 as curve (a) which was obtained at the atomic density $N = 1.3 \times 10^{17}$ cm$^{-3}$. The dipole–dipole interaction and collision broadening dominate at this atomic density, the spectral width due to self-broadening is larger than the ground state hyperfine splitting and the ground state hyperfine structures in reflectivity spectra that can be seen at low atomic density are not resolved. While we apply the pump laser beam, atoms are partially excited and the dipole–dipole interaction is reduced. Thus, the self-broadened line width is also reduced [8–11], and the ground state hyperfine structures start to be revealed as described in [9, 10]. The narrowed FM spectrum with pump power $P = 180$ mW is shown as curve (b) in figure 3.
As described in [10], the reflection spectra as well as the FM reflection spectra can be interpreted in terms of the dielectric coefficient of atomic vapour. Taking into account the excitation, the dielectric coefficient $\epsilon$ of a two-level atomic system is given as

$$\epsilon(\omega) = 1 + \frac{k\eta N}{\Delta\omega + \Delta \Omega - i\Gamma},$$

(1)

where $N$ is the atomic density, $\Delta\omega$ is the frequency detuning, $\Delta \Omega$ is the overall line shift which includes Lorentz and non-Lorentz shift [12–14] and $\Gamma$ is the self-broadened linewidth. The constant $k$ is given by $k = f c r_e \lambda$, where $f$ the oscillator strength of transition, $r_e$ the classical radius of electron, $\lambda$ the wavelength of transition and $c$ is the speed of light in vacuum.

An excitation factor $\eta$ is defined as the fractional population difference between ground and excited states

$$\eta = \frac{N_g - N_e g_e / g_g}{N},$$

(2)

where $N_g$ and $N_e$ are the ground and excited state atomic densities respectively, $g_e$ and $g_g$ are the degeneracies of the ground and excited states, respectively. Maximum excitation corresponds to $\eta = 0$ and zero excitation $\eta = 1$. Using this expression for the dielectric coefficient $\epsilon$ and Fresnel formula, we are able to calculate the reflectivity and FM spectra which is the derivative of reflectivity with respect to frequency.

In order to obtain the width and excitation factor from the experimental data, we use the expression of FM spectra to fit the experiment data by leaving the self-broadened width $\Gamma$, the excitation factor $\eta$ and the line shift $\Delta \Omega$ as fitting parameters. The excitation factor $\eta$ is normalized to unity for the case where no pump laser is applied. In our experiment, Rb vapour contains natural abundance of $^{85}$Rb and $^{87}$Rb which gives rise to four Doppler-broadened absorption lines in the absorption spectra. All of four components are taken into account in the fitting of reflection spectra. Each component is given a normalized oscillator strength. The dashed curves in figure 3 are the examples of the fitting. For the case without the pump laser, the fitted width is $13.0 \pm 0.3$ GHz and $\eta = 1.0$; for the case with the pump laser (laser power $P = 180$ mW), the fitted width is $4.98 \pm 0.05$ GHz and $\eta = 0.36$. At the same atomic density ($N = 1.3 \times 10^{17}$ cm$^{-3}$), the FM spectrum is recorded when we apply the pump laser with different powers. The fitting of these FM spectra gives the widths corresponding to the different excitation factors. The fitting results are shown as the red squares in figure 4, where the width is plotted as a function of the excitation factor $\eta$. The dashed line in figure 4 is a linear fit ($y = a + bx$, where the dependent variable $x$ represents the excitation $\eta$ and $a$ and $b$ are fitting parameters) of the excitation dependence of the width at atomic density $N = 1.3 \times 10^{17}$ cm$^{-3}$, and the slope (fitting parameter $b$) is $12.7$ GHz.

We determine the widths and excitation factors by measuring and fitting the FM reflection spectra at different atomic densities from $N = 1.3 \times 10^{17}$ cm$^{-3}$ to $N = 2.2 \times 10^{16}$ cm$^{-3}$. The solid straight line is a linear fit.
$10^{16}$ cm$^{-3}$. The measured density dependence of the self-broadened rubidium $D_2$ linewidth is the same as in [15]. The excitation dependence of the width for different atomic densities is shown in figure 4 with different colours. The corresponding linear fit gives the slope (width/$\eta$) for each density. In figure 5, the slope is plotted as a function of the atomic density. The solid line is a linear fit. If the slope for each density is normalized by taking the ratio of the slope to the width without the pump laser at each density, the normalized slopes are close to unity. As shown in figure 6, the value of the normalized slope is $0.90 \pm 0.05$. According to our simple model for the fit, the normalized slope is density independent. It indicates that the self-broadening at high atomic density is a combination of collision and static spectral broadening (inhomogeneous profile).

3. Conclusion

We have observed that the excitation dependence of self-broadening is the same in the range of atomic density from $2.2 \times 10^{16}$ to $1.3 \times 10^{17}$ cm$^{-3}$, where dipole–dipole interactions are a dominant source of spectral broadening. Our results support the predictions of the theoretical model developed in [4]. The obtained results can be useful for understanding of excitation processes in a condensed media such as solutions, glasses, polymers, proteins and molecular crystals. In atomic gases it will be interesting to study a possible transition from many body interactions (disordered excitons) [4, 7, 8] to the resonance two-body interaction (impact collisions) at lower atomic densities [1, 2]. Probably the studies will require applications of nonlinear optical methods in frequency and time domain as the complimentary spectroscopic techniques. We shall note that additional information about the dipole–dipole interaction in a dense gas may be obtained by using a nano-cell [16].

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