

Experimental study of power broadening in a two-level atom*

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Two frequency-stabilized cw dye lasers and an atomic beam were used to prepare sodium atoms in the aligned $F = 2$, $M_F = 2$ ground state, and to study power broadening in the transition from this ground state to the $3P_{3/2}$ $F = 3$, $M_F = 3$ excited state. This system is, to extremely good accuracy, a two-level atom. The line shape was determined as a function of the incident field strength and found to be accurately a Lorentzian over a large range of incident field strengths. The power broadening and saturation at resonance of the two-level system also agree with theory.

I. INTRODUCTION

In this paper, we will describe a series of experiments studying the natural line shape of a dipole transition in an isolated atom and the way in which the line is broadened by intense applied fields. Power broadening has been observed many times, but here we will study the effect in a very simple system under precisely controlled conditions. The two-level atom is one of the most extensively studied models in theoretical physics. It has provided a useful framework to describe much of the work which has been carried out in resonant quantum optics for the past ten years. In spite of this, atoms always have many levels and the two-level atom is usually only a rough model which ignores underlying details that are not resolved in the given experiment.

Because the theory is so well understood,¹ and can be worked out even from first-principle quantum electrodynamics, it is desirable to carry out an experiment for which the two-level atom is really an accurate model. In that case, the theory and experiment can be compared without the ambiguities and approximations that usually obscure such comparisons in more complex systems.

We have prepared this simple two-level atom by using a frequency-stabilized dye laser to optically pump sodium atoms in an atomic beam into a particular Zeeman sublevel of one hyperfine component of the ground state. This level is coupled by a second laser beam to one and only one Zeeman component of a hyperfine component of an excited state. The rate of absorption of this second laser beam is measured as a function of its frequency and intensity. The results are found to be in very good agreement with theory.

II. PREPARATION OF A TWO-LEVEL ATOM

One particular transition between two hyperfine components of the D_2 line in sodium has some rather special properties. This transition is be-

tween the $3S_{1/2}$ $F = 2$, $M_F = 2$ ground state and the $3P_{3/2}$ $F = 3$, $M_F = 3$ excited state. Each of these states is an aligned state in which the magnetic moments associated with the nucleus, the electron's spin, and its orbital angular momentum are aligned. A laser beam propagating along the quantization axis and circularly polarized (σ^+ polarized) will excite an atom in this ground state to this one particular excited state, and not to any other sublevel of the entire $3P$ manifold. Furthermore, an atom in this excited state can decay by a dipole transition into only this one ground state. These states are shown in Fig. 1. This alignment effectively eliminates the hyperfine and Zeeman splittings and makes a simple two-level atom. The problem is then to prepare the atom in this particular ground state.

Abate² has described a technique by which a dye laser can be used to optically pump the atom into the aligned state. Walther³ and Wu *et al.*⁴ have used this technique to study the emission spectrum of a two-level system. The technique is most easily understood by referring to Fig. 2. The Zeeman components of the $F = 2$ ground state and the $F = 3$ excited state are shown. In the absence of a magnetic field, the sublevels are degenerate. A σ^+ polarized laser beam will cause $\Delta M_F = +1$ transitions on absorption (solid lines), while spontaneous emission back to the ground state allows $\Delta M_F = \pm 1, 0$ (dotted lines). If an atom spends a hundred radiative lifetimes in such a laser field, it will repeat these processes a hundred times and be very accurately pumped into the aligned state. Angular momentum is absorbed from the polarized field until the atom reaches its state of maximum angular momentum.

Of course, it is difficult to prepare perfectly circularly polarized light. Further, if there is no magnetic field along the quantization axis, then the slightest component of stray field orthogonal to the quantization axis will cause the aligned spins to precess, mixing the population into other levels. To counteract this in our experiment, we have ap-

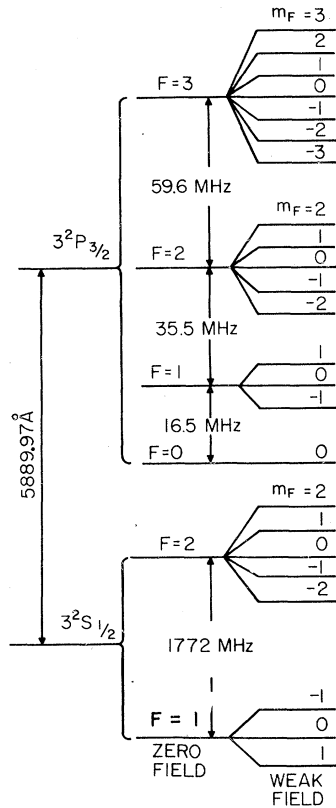


FIG. 1. Hyperfine structure of the sodium D_2 line. The splitting of the ground state is sufficiently large so that transitions from only one of the sublevels are resonant at a time.

plied a magnetic field of about 1 G along the quantization axis. This splits the degeneracy by an amount small compared to the natural linewidth. It does, however, assure that the aligned state will be maintained even in the presence of stray

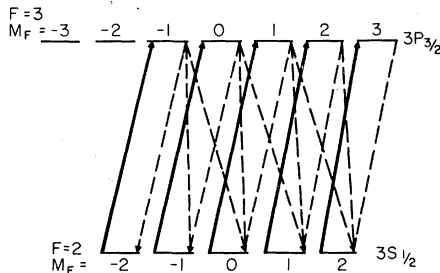


FIG. 2. Optical pumping the atoms into an aligned state. The atoms scatter about 100 photons in passing through the laser beam. The heavy solid lines represent stimulated absorption of the circularly polarized laser beam, while the dotted lines represent the mixed polarizations of the spontaneous emissions. The population rapidly accumulates in the $m_F = 2$ ground state.

fields since the total magnetic field will be nearly along the quantization axis.

III. POWER BROADENING THEORY

The isolated atoms in the atomic beam are influenced by only one damping and broadening mechanism: spontaneous emission. There has been a great deal of discussion in the past few years concerning the adequacy of various semiclassical theories in describing such a system.⁵ Recently, however, it has been shown that the Heisenberg equations of motion in quantum electrodynamics describing an atom interacting with the quantized field which is initially in a coherent state, give single time expectation values satisfying the semiclassical optical Bloch equations (OBE).⁶ Thus, conventional semiclassical theory and quantum electrodynamics give precisely the same predictions for our experiments. Neoclassical theory does not treat spontaneous emission the same way, and gives different results.

In these experiments, the atoms spend at least a hundred radiative lifetimes in the laser beam so that they are effectively in a steady-state condition throughout the experiments. If one takes the steady-state solutions to the OBE and calculates the rate of absorption of energy from the field one finds

$$I(\Delta) = -(\hbar\omega/2)\kappa\epsilon v(\infty, \Delta) \\ = \frac{\hbar\omega}{2T} \frac{(\kappa\epsilon)^2}{(\kappa\epsilon)^2 + 1/(2T^2) + 2\Delta^2} \quad (3.1)$$

Here we have used conventional definitions: $\kappa\epsilon$ is the Rabi frequency, Δ is the detuning of the applied field frequency from the atomic resonance, $T = 1/A$ with A being the Einstein A coefficient, and $v(\infty, \Delta)$ is the steady-state value of the out-of-phase component of the atomic dipole moment.¹

The absorption spectrum is then a Lorentzian with a full width at half maximum (FWHM) of

$$\gamma_\omega = (1/T)[1 + 2(\kappa\epsilon)^2 T^2]^{1/2} \quad (3.2)$$

At low powers, the FWHM is the natural linewidth $1/T$, but as the power is increased, the spectrum is power broadened. Also, as the power is increased the rate of absorption at resonance, $I(\Delta = 0)$, approaches $\hbar\omega/2T$ and saturates.

For comparison with experiment, it is convenient to introduce a dimensionless intensity

$$I_D = 2T^2(\kappa\epsilon)^2 = I_{\text{lab}}/(6 \text{ mW/cm}^2) \quad (3.3)$$

Then, the absorption spectrum can be written

$$I(\Delta) = \frac{\hbar\omega}{2T} \frac{I_D/4T^2}{\Delta^2 + (I_D + 1)/4T^2} \quad (3.4)$$

and the FWHM of the Lorentzian is

$$\gamma_\omega = (1/T)(I_D + 1)^{1/2}. \quad (3.5)$$

The saturation is best characterized by the dimensionless quantity

$$\begin{aligned} S_R &= (2T/\hbar\omega)I(\Delta=0) \\ &= I_D/(I_D + 1). \end{aligned} \quad (3.6)$$

We will refer to this as the relative saturation. Since frequency is measured in the laboratory in units of Hz, it is convenient to write (3.5) in the form^{7,8}

$$\gamma_\nu = (10 \text{ MHz}) \times (I_D + 1)^{1/2}. \quad (3.7)$$

IV. EXPERIMENTAL APPARATUS AND PROCEDURE

The experimental apparatus is shown schematically in Fig. 3. A thermal sodium atomic beam is collimated by two pinholes to a 2.5-mrad maximum divergence angle. The atoms traveling with a mean velocity of 9×10^4 cm/sec pass through a laser beam which is 2 mm in diameter (e^{-2} points) and has a Gaussian profile. The laser is locked in frequency by an electronic servo-system to the $F=2 \rightarrow 3$ transition, and is stable to within ± 2 MHz and provides about 50 mW/cm². The atoms spend about 140 radiative lifetimes in passing through this beam and in excess of 95% of the population is pumped into the $M_F=2$ sublevel of the ground state. This population distribution was verified by observing the polarization of the scattered light, and by noting the absence of the $F=2 \rightarrow 2$ transition in the absorption spectrum.

The atoms then enter the field of the scanning laser. This field is a 3-mm-wide Gaussian centered 5 mm from the center of the pumping beam. The interaction region is imaged without magnification onto a 0.5-mm pinhole, which can be

translated along the atomic beam. In this way, the light scattered from a small subregion, over which the intensity varies only slightly, can be measured. The scanning laser is locked by an electronic servo-system to the maximum of a passband of a thermally isolated scanning Fabry-Perot interferometer. This interferometer is then tuned through the atomic resonance, and the laser follows with its frequency jitter limited to ± 1.0 MHz. Long-term drift of the interferometer did not affect the experiment. The limit on short-term drift was established by rapidly scanning the laser a number of times through a narrow resonance and verifying that the traces fell on top of each other. Three mutually perpendicular sets of Helmholtz coils, that are about the vacuum chamber, cancel the Earth's field and provide the magnetic field in the z direction to prevent remixing. The alignment requirements are rather stringent. The two laser beams must be parallel to each other and perpendicular to the atomic beam to within a few milliradians. The magnetic field must be parallel to the laser beams to about the same accuracy. The resonant interaction itself proved to be the most accurate measure of alignment. In order to adjust the laser beams orthogonal to the atomic beam, a corner cube is used to reflect one of the laser beams back through the atomic beam parallel to itself but displaced by a few millimeters. Then, if the incident laser beam makes an acute angle with the velocity vector of the atoms, the reflected beam will make an obtuse angle. One of the beams will be Doppler shifted upward in frequency and the other downward. If the laser is then scanned in frequency through resonance and the fluorescence from the two interaction regions monitored, perfect alignment will be achieved when the incident and reflected beams are simultaneously resonant. This method allows a quick, reliable way of achieving alignment which reduces Doppler broadening to the limits imposed by the beam divergence. The magnetic field direction and laser polarization were adjusted to approximately their optimum values by conventional methods, and then the fine adjustments were made spectroscopically. This was done by adjusting the magnetic field and the retardation of the Soleil-Babinet compensator to achieve the most efficient possible optical pumping. This was determined by scanning the laser through the $F=2 \rightarrow 2$ and $F=2 \rightarrow 3$ transitions and adjusting the system so that there was no trace of the $F=2 \rightarrow 2$ resonance at low powers. In this way, birefringence of the vacuum chamber windows and magnetic field inhomogeneities could be compensated for directly.

After aligning the system, the absorption spectra were determined at a series of different incident

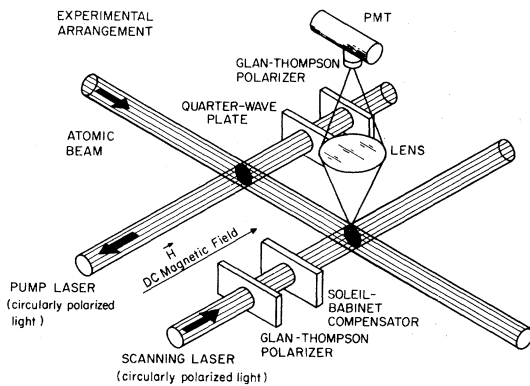


FIG. 3. Experimental arrangement. The atomic beam passes through a first laser beam, which prepares the two-level atom, and then through a second laser beam which is scanned in frequency determining the absorption spectrum.

field strengths. The scanning laser intensity was adjusted with a variable density filter. At each intensity level, the photomultiplier tube on the top was adjusted to correct for small shifts in the position of the laser beam. Each absorption spectrum was measured five times, digitized and averaged. In addition, the atomic beam was blocked off before the last collimating aperture and an additional scan of the laser then allowed an accurate measure of the background scattering. This was especially important because of the slow fall-off of the Lorentzian tails.

The intensity of the scanning laser beam was measured with a United Detector Technology power meter calibrated to $\pm 7\%$. A 1-mm^2 pinhole was placed in front of the detector, and they were translated back and forth to find the peak of the Gaussian profile. The stated intensities are averages over this aperture.

The remaining principal sources of error were residual divergence of the atomic beam, nonuniformity of laser intensity due to diffraction of the laser beam from apertures in the system, and noise in the detection system. All of these effects have been estimated and fall within the experimental error bars given with the data.

V. RESULTS

The five absorption curves taken at each intensity level are averaged together, and the averaged curves are fitted with a Lorentzian by using the method of least squares. The zero point for scattered intensity is corrected for background scattering, and the origin of frequency is at the line of symmetry. The error estimate is the 95% confidence interval of the fit of the data to the Lorentzian. The frequency scale is determined by measuring the distance between the $F=3 \rightarrow 2$ and $F=2 \rightarrow 2$ transition when the atoms are not prepumped and the scanning laser's intensity is low. This is known to be 59.6 MHz.

The experimentally measured absorption spectra are shown in Fig. 4. Each curve is normalized to unit intensity at resonance. The dots represent the experimental absorption spectrum and the solid curve is a best Lorentzian fit. In Fig. 4(a), the absorption spectrum is for a low-intensity case, in which the laser irradiance is 0.84 mW/cm^2 . The FWHM is 12.4 MHz due to power broadening and the 1.5-MHz residual Doppler broadening of the atomic beam. In Fig. 4(b), the laser intensity is 3.5 mW/cm^2 . This is the intensity required to begin saturation, i.e., the intensity such that spontaneous and stimulated emission are equally likely. In Fig. 4(c), a high-intensity case, 90 mW/cm^2 , is given. The $F=2 \rightarrow 2$ transition which would be pres-

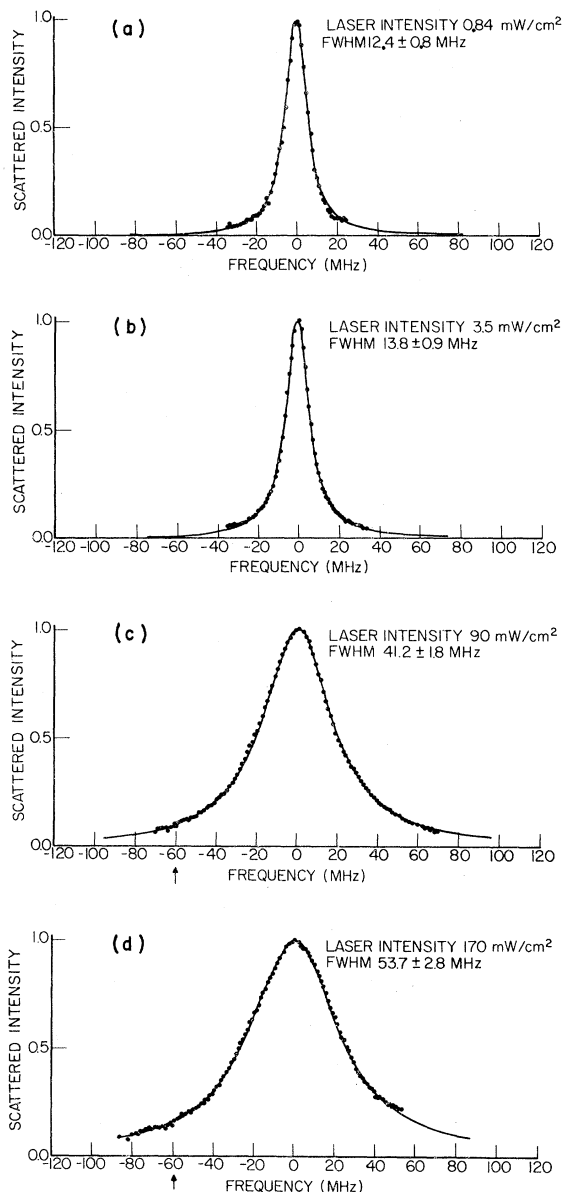


FIG. 4. Measured absorption spectra. The dots are experimental points averaged over five laser scans, the solid curves are best fit Lorentzians. The arrow on the frequency scale points to the location of the missing $F=2 \rightarrow 2$ transition. The stated error is the 95% confidence interval.

ent in an unpumped atom would be at -59.6 MHz as is indicated by the arrow. There is no sign of the $F=2 \rightarrow 2$ transition, and the curves are all to an extremely good precision Lorentzian. Figure 4(d) shows an even higher intensity level of 170 mW/cm^2 .

We compare the experimental widths to the theoretical predictions for power broadening in Fig. 5. We have taken the FWHM of the best-fit

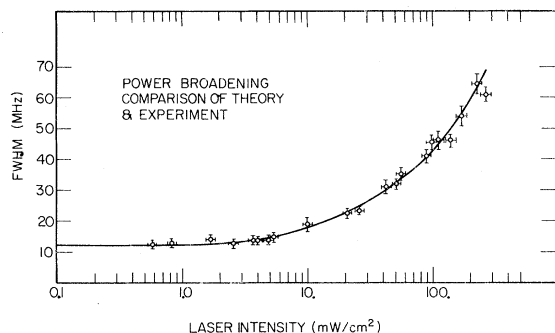


FIG. 5. Comparison of measured power broadening and theoretical prediction. The widths of the best-fitting Lorentzians are compared with theory. There are no free parameters.

Lorentzians of our line-shape measurement and plotted them as a function of the laser intensity. The solid curve is the theory (QED plus 1.5-MHz Doppler broadening) with no adjustable parameters. Again, agreement between theory and experiment is very good. The intensity scale is logarithmic, so that the range of intensities is from 0.5 mW/cm² to about 300 mW/cm². Note that the power-broadened line greatly overlaps the other transition at the high powers. The error bars on the experimental points are calculated from the error for the curve fit of our Lorentzians and from the accuracy of the calibration of the intensity measuring device.

In Fig. 6, we display the saturation of the absorption rate at resonance, S_R . There is a scaling parameter that depends on the number of atoms scattering, the initial population of the two-level atom, and the detection geometry. This parameter is chosen but once, and all of the data in a given experiment multiplied by it. The data shown in the saturation curve were taken on a number of different occasions, and on each occasion the full intensity range was covered. In this figure, the solid line is theoretical and the error bars are defined as in the previous graph. Again, our two-level theory is quite adequate.

The experimental data for saturation and power broadening were not taken above 300 mW/cm² because the remaining unavoidable small misalign-

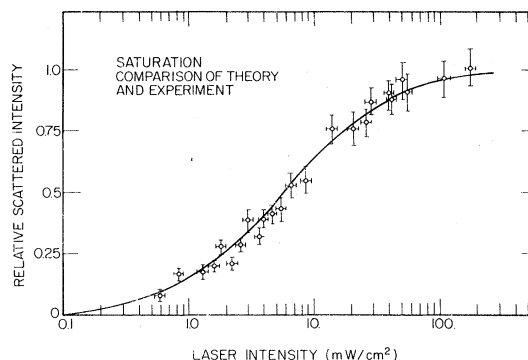


FIG. 6. Comparison of observed resonant saturation with theory. The scattered intensity at exact resonance is plotted as a function of the laser intensity. The data is scaled with a parameter dependent on the number of atoms and the detection efficiency.

ments of the circular polarizer and magnetic field made it impossible to maintain the population in the two-level system. At this power level an ellipticity in the polarization corresponding to 2% of the laser intensity going into the σ^- component would saturate the transitions pumping out of the two-level system. These effects were studied and are described elsewhere.⁹

VI. DISCUSSION

By optical pumping, we have prepared the atoms in a sodium atomic beam in an aligned state which is to good approximation an isolated two-level atom. We have carried out a series of experiments studying the natural line shape and the way in which it power broadens. The results are in very good agreement with quantum-electrodynamic theoretical predictions. The absorption line shape was accurately Lorentzian with the proper width over $2\frac{1}{2}$ orders of magnitude variation in incident field intensity. The scattered intensity saturated according to the theoretical predictions.

Some aspects of these effects have been seen previously in more complicated systems, but in this series of experiments we have taken care to produce a system in which theory and experiment can be accurately compared. We find that the theory and experiment are in good agreement.

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