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Subnatural optical pumping dips

C. M. Klimcak, J. C. Camparo, R. A. Cook, and R. P. Frueholz
Chemistry and Physics Laboratory, The Aerospace Corporation, P.O. Box 92957, Los Angeles, California 90009
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The technique of optical pumping dips has been employed to generate subnatural structure in a ^{87}Rb D_1 line (natural width 5.6 MHz). Structure as narrow as 1.4 MHz was observed. These experiments represent the first observation of subnatural spectral structure generated without requiring atomic coherence. Additionally, line shapes observed in these studies may be analyzed to yield transition dipole moments without requiring internal standards or an absolute calibration of fluorescence collection efficiency in the experimental apparatus.

In recent years there has been a growing interest in spectroscopic techniques capable of reducing the linewidth of an optical transition below that specified by spontaneous emission. Resonances narrower than the natural linewidth have been observed via optical rf double-resonance experiments,[1,2] level-crossing studies,[3,4] quantum-beat experiments,[5] and optical phase-switching techniques.[6] Additionally, optical Ramsey experiments have the potential to yield subnatural structure.[7–9] In contrast to the above studies where transition line shapes were narrowed upon application of the various techniques, Gawlik *et al.*[10] observed a dip, with a subnatural width, in a homogeneous line through the use of polarization spectroscopy. Thus far, in all examples of subnatural spectroscopy, the observed features have been the result of coherent interactions induced by the exciting fields.[11] In this paper we experimentally demonstrate for the first time that subnatural structure may also be generated using a technique which is not based on coherent interactions. The present technique, utilizing optical pumping, and its potential for producing subnatural structure were first discussed by Klimcak and Camparo[12] who used it to generate dips in a homogeneous optical line shape. Similar dips were observed subsequently by Cerez *et al.*[13] studying optical pumping in a cesium atomic beam. We now show that these optical pumping dips may have widths narrower than the natural linewidth (5.6 MHz) of a hyperfine component of the ^{87}Rb D_1 line.

In the present experiment, as shown schematically in Fig. 1(a), the output of an actively stabilized single-mode cw dye laser is split and directed transversely through two regions of an effusive Rb atomic beam. The atomic beam has been collimated such that its transverse Doppler width is less than the natural linewidth of the transition under study. Atoms in the beam interact sequentially with both laser fields. As indicated in Fig. 1(b), the laser frequency is tuned across the $5\,^2P_{1/2}(F=2)$–$5\,^2S_{1/2}(F=1)$ D_1 transition at 794.7 nm. The primary action of the laser in the first interaction region is to reduce the population of the $5\,^2S_{1/2}(F=1)$ state through optical pumping into the $5\,^2S_{1/2}(F=2)$ state. The detected signal is the fluorescence produced in the second region as the frequency of the laser illuminating both regions is scanned across the atomic resonance. When the laser intensity in each region is appropriately adjusted the fluorescence signal assumes the theoretical form shown in Fig. 1(c). Essentially, a dip is generated in the center of the fluorescence line shape. The dip is the result of the enhanced optical pumping efficiency of the center of the absorption line.

In the present experiments the atomic beam source is held at 473 K and the interaction regions are separated by 55 cm yielding an average transit time greater than 1 msec. Thus any coherence generated in the first interaction region has decayed by the time the atoms reach the

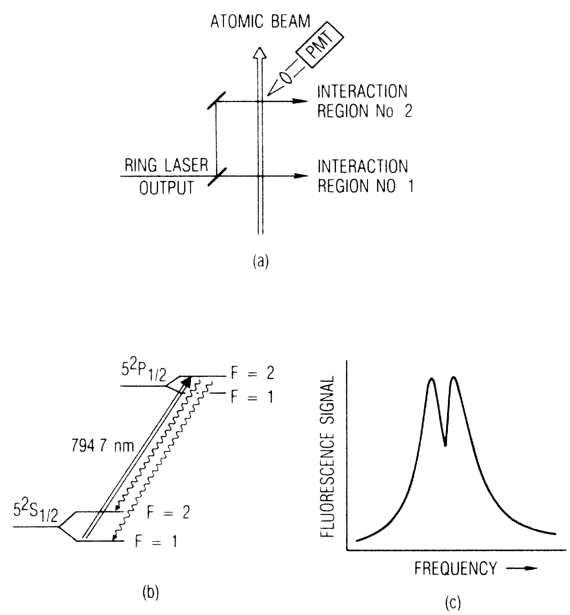

FIG. 1. (a) Schematic diagram of experimental configuration. (b) ^{87}Rb energy-level diagram showing radiative processes of interest. The straight line represents laser excitation between the $5\,^2P_{1/2}(F=2)$ and $5\,^2S_{1/2}(F=1)$ levels as well as stimulated emission. Optical pumping occurs because of the $5\,^2S_{1/2}(F=2)$ fluorescence decay channel. (c) Typical optical pumping dip line shape.

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second interaction region. The distance over which atoms interact with the laser in the first region is 1.3 cm and 0.16 cm in the second region. All line shapes presented are the result of 10-min scans of the cw ring laser's (Spectra-Physics 380 D) frequency. Photon counting is employed for the detection of the fluorescence signal. In Fig. 2 the experimental line shapes for a range of laser irradiances in the first interaction region and a fixed laser irradiance 2 mW/cm^2 in the second interaction region are displayed. When the laser beam entering the first interaction region is blocked, the curve (a) in Fig. 2 is observed. The linewidth, 27 MHz, is well above the 5.6 MHz natural linewidth and is due to optical pumping and power broadening in the second region. For a laser irradiance in region 1 of 0.5 $\mu W/cm^2$, curve (b), which shows a dip with a linewidth of 11 MHz, is obtained. As the irradiance in region 1 is reduced the depth and width of the dip are observed to decrease. For curves (c) and (d) with irradiances of approximately 0.06 and 0.05 $\mu W/cm^2$, respectively, only the central portion of the transition line shapes are displayed with dip widths of 3.3 and 1.4 MHz being observed respectively. The widths of the dips are subnatural with the narrowest feature approximately one-quarter the natural width. The decrease in the depth of the dip is quite significant with that of curve (d) being only 5% that of the dip in curve (b).

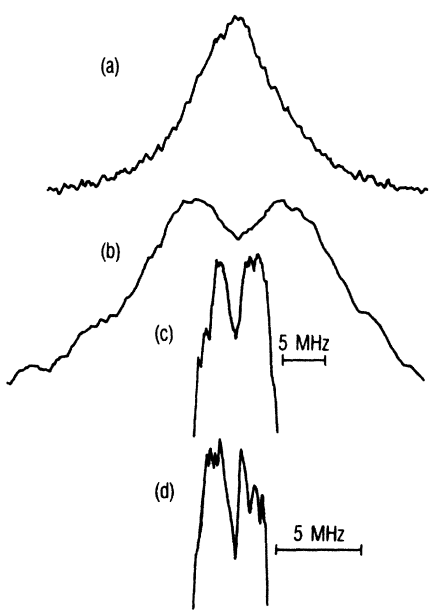

FIG. 2. Experimental line shapes resulting from detection of laser-induced fluorescence in interaction region 2. For all curves laser irradiance in region 2 was 2 mW/cm^2. (a) Laser into interaction region 1 blocked. For remaining curves laser irradiance and dip (full width at half maximum) are given. (b) 0.50 $\mu W/cm^2$, 11 MHz, (c) 0.06 $\mu W/cm^2$, 3.3 MHz, (d) 0.05 $\mu W/cm^2$, 1.4 MHz. For clarity the horizontal frequency scale used to display curve (d) has been expanded by a factor of 2 compared to that used for curves (a), (b), and (c). Curves (c) and (d) both display subnatural optical-pumping dips. The ratio of the depth of the dip to peak height seen in (d) is 5% of that observed in (b).

Previously it was found that the optical pumping dip line shape could be generated by treating the ^{87}Rb atom as a three-level system.[12] As indicated in Fig. 1(b) the three levels considered are the two hyperfine levels of the ^{87}Rb $5\,^2S_{1/2}$ ground state along with the $F=2$ hyperfine level of the $5\,^2P_{1/2}$ fine-structure component. In order to describe the dip mechanism, it is only necessary to apply a rate-equation analysis to the level population densities in each of the interaction regions. The initial conditions for the rate equations of the second region are just the level population densities of the atoms exiting the first interaction region. Calculated line shapes presented in this paper were obtained in the same manner as previously[12] except for the following aspects. Intensities in the second interaction region are sufficient to require that stimulated-emission terms be incorporated into the rate equations. Also, line shapes have been averaged over the velocities in a thermal atomic beam.

Within the simple three-level model the processes leading to the observed line shapes are easily understood. The fluorescence signal detected in the second interaction region is dependent on both the population density of the lower level interacting with the laser as well as the rate at which photons are absorbed by atoms in that level. As either quantity is increased the detected fluorescence will also increase. When the laser frequency in both regions is scanned across the optical resonance the fluorescence signal depends on laser detuning in two ways. As the laser frequency in the second region approaches resonance the fluorescence signal will attempt to increase as a result of the increased rate of photon absorption. Simultaneously, the increasing rate of optical pumping in the first region reduces the population density of the probed level. These competing processes distort the line shape and for appropriate laser intensities yield optical pumping dips.

The rate-equation analysis is able to reproduce the experimental results reasonably well. From this analysis we find that if the laser beams in both interaction regions are parallel, the minimum in the dip will occur at the line center. Due to the small depths of narrow dips, though, it is not clear that the dip center will provide a more precise determination of a resonance frequency than could be obtained by directly analyzing the transition's natural line shape. We also note that the only parameter in the analysis of these line shapes, which is not easily measured during the course of the experiment, is the transition dipole moment of the optical resonance. Consequently, matching a calculated line shape with the experimentally obtained parameters and line shape should provide a straightforward means of determining this last quantity. Thus for atomic transitions and potentially molecular transitions which can be optically pumped, this technique provides a novel means of measuring transition dipole strengths. The potential of this technique may be illustrated using the experimentally measured parameters that yielded curve (b) of Fig. 2. Inserting these values along with the known transition dipole moment of this resonance, 2.5×10^{-29} C m (Refs. 14 and 15), into the rate-equation analysis yields curve (a) in Fig. 3, which is the upper portion of the entire line shape. Varying the transition dipole moment used in the calculation by $\pm25\%$

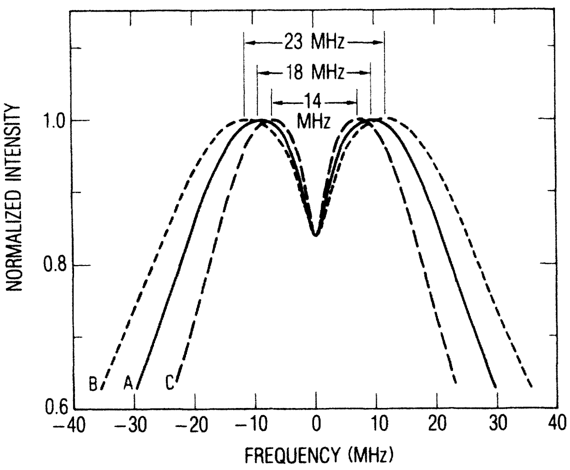

FIG. 3. Optical-pumping-dip line shapes resulting from rate-equation analysis using experimentally measured parameters and varying the magnitude of the transition dipole moment. Transition dipole moment values employed: (a) 2.5×10^{-29} C m (correct value), (b) 3.0×10^{-29} C m (+ 25%), and (c) 1.9×10^{-29} C m (−25%). The maximum amplitudes of the line shapes have been normalized to the same value. The frequency spacing between the two peaks generated by the dip is seen to be particularly sensitive to transition dipole moment variation.

yields line shapes (b) and (c), respectively, in Fig. 3. The maximum amplitudes of each of the curves have been normalized to the same value. It appears that the dip features, particularly the frequency interval between the two peaks generated by the dip, are sensitive enough to allow the transition dipole moment to be determined to better than 25%.

As a concluding remark we note that it is not necessary to employ two physically isolated beams to generate optical pumping dips. A single beam appropriately apertured could yield two excitation regions separated by a sufficient distance to allow any coherence to decay. The configuration may be useful when observing dip structure as any lack of parallelism between the two physically separated beams can result in a asymmetry in the optical pumping dip line shape as well as a decreased dip depth.[12]

Briefly summarizing, in this paper we demonstrate that optical-pumping dips in homogeneously broadened lines can display subnatural widths. The observed subnatural features are the first generated without application of a coherent phenomena. We also indicate that the experimental line shapes along with a straightforward rate-equation analysis provide a new means of extracting transition dipole moments. Finally, while the experiment is typically configured with two physically separated laser paths this could be replaced by a single appropriately apertured laser beam, reducing experimental alignment requirements.

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