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LETTER TO THE EDITOR

Observation of the resonant Stark effect at optical frequencies†

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Abstract. The spectrum of resonantly scattered light at right angles to a sodium atomic beam is reported. The light source was a CW dye laser tuned to resonance with a hyperfine component of the D_2 line, and incident at right angles to the atomic beam. The spectrum, with the Stark effect sidebands, was recorded as a function of both the laser intensity and its detuning from resonance. The overall resolution is better than 20 MHz.

In recent years there have been a large number of attempts to predict the spectrum of the light scattered by an isolated atom illuminated by a monochromatic field tuned accurately to resonance with a two-level transition. These theoretical calculations have variously predicted a sharp line scattered spectrum (Heitler 1954), a lorentzian with a hole burned in the middle (Bergmann 1967, Chang and Stehle 1971), a three peaked spectrum (Apanasevich 1964, Newstein 1968, Mollow 1969, Stroud 1971, 1973), and even more complex shapes (Morozov 1969, Gush and Gush 1972). There have been some experimental studies in this area (Hertz *et al* 1968, Hänsch *et al* 1969, Shahin and Hänsch 1973), but none have yet had the resolution to provide an unambiguous answer to this problem. In this letter we will describe an experiment which measures this spectrum in detail and determines that over a wide range of incident field intensities, the spectrum has three peaks.

This structure in the scattered spectrum has been termed the dynamic, or AC, Stark effect. The Stark effect produced by an optical field is dramatically enhanced by tuning the field to within the natural linewidth of a two level transition. We have taken advantage of the precise tunability of a dye laser, as well as the well defined resonance frequency of a collimated atomic beam illuminated at right angles, to maximize the observed effect and make detailed measurements possible.

The usual heuristic explanation of the dynamic Stark effect is that it is just the analog in frequency space of optical nutation. We have found, however, that we are unable to explain our experiments in terms of the conventional phenomenologically damped semiclassical equations usually used to describe nutation (Tang and Silverman 1966, Hocker and Tang 1968, Hoff *et al* 1970, Brewer and Shoemaker 1973). Thus the present experiment serves to point up limitations of phenomenological semiclassical theory as well as determining the nature of the spectrum.

The apparatus for observation of the resonant effect at 5890 Å in sodium is similar to that described in an earlier publication (Schuda *et al* 1973). A dye laser beam and an

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atomic beam are crossed at right angles and the resonant fluorescence from the interaction region is analysed. Figure 1 shows the experimental set-up including intensity monitoring apparatus and Fabry–Perot interferometer.

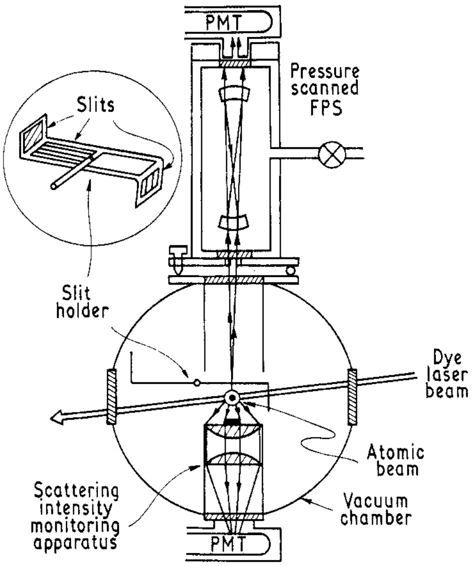

Figure 1. Experimental apparatus for AC Stark effect observation. The atomic beam is travelling out of the plane of the paper.

The CW dye laser is first coarse-tuned to resonance in a sodium absorption cell. The laser is then directed to intersect the atomic beam and fine-tuned to the hyperfine transition $F = 2 \rightarrow 3$ of the sodium D_2 line by monitoring the resonance scattering from the interaction region. This particular atomic transition was chosen because it is a two-level transition and the atomic population cannot be optically pumped into a third level (see figure 2*a*). The dye laser is stable to within $\pm 1{\cdot}5$ MHz for a period of 10 seconds so that during a single 5 s spectral scan the laser frequency remains essentially stationary relative to the 10 MHz natural linewidth.

Upon entering the chamber the laser beam is apertured by a slit shown in figure 1. The data presented here was taken with the 1 mm wide slit so that the atoms coming down the atomic beam are illuminated over a 1 mm region; the corresponding transit time is approximately 100 natural lifetimes. The laser beam electric field vector is parallel to the atomic beam. The atomic beam is collimated to 4 milliradians by passing through two 0·5 mm circular apertures which are 25 cm apart. (The reason for the relatively large apertures is to obtain a large atomic flux and therefore a large scattered intensity.) The dye laser and atomic beam are perpendicular to within 2 milliradians, so that the residual Doppler width in excitation is no more than 5 MHz.

Scattered light from the interaction region can go into one of two collecting systems. The scattered intensity monitoring system merely collects the scattered light within a large solid angle and indicates the location of the laser frequency relative to the centre of the absorption line. The distance from the centre frequency is determined from our knowledge of the relative scattering cross section as a function ofinput power and detuning.

Light scattered from the atomic beam also enters the confocal Fabry–Perot interferometer on the top of the vacuum chamber. A blackened piece of foil covers the central area of the intensity-monitoring lenses so that stray light is not reflected into the Fabry–Perot. The interferometer is followed by an aperture which allows only light scattered at right angles to the atomic beam to be detected, while at the same time

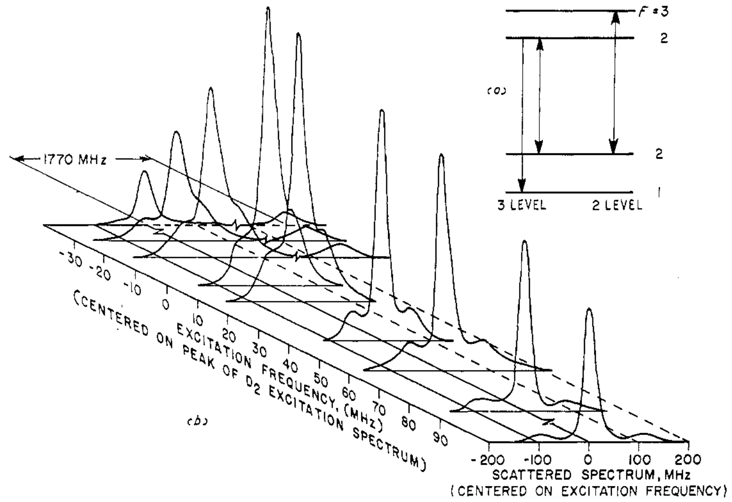

Figure 2. (*a*) The D_2 transitions excited by the dye laser. $F = 3$ and $F = 2$ are hyperfine components of the $3^2P_{3/2}$ level and are separated by 60 MHz. $F = 2$ and $F = 1$ of the ground $3^2S_{1/2}$ level are separated by 1772 MHz. The 2 and 3 level systems arise from the $\Delta F = 0, \pm 1$ selection rules. (*b*) Composite graph of the spectra of the scattered light for various laser detunings. The laser intensity is 406 mW cm^{-2} $\pm$ 100 mW cm^{-2}. The measurements of excitation frequency are accurate to within ± 7 MHz, and the scale on the scattered spectra is accurate to ± 10 MHz.

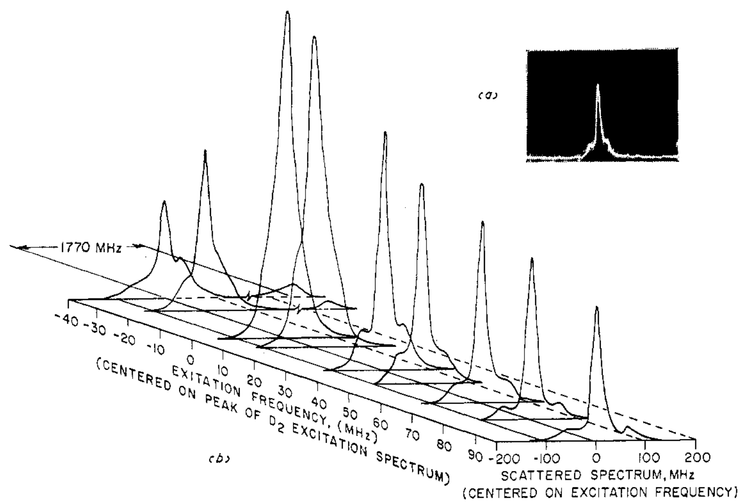

Figure 3. (*a*) Typical oscilloscope trace of emission spectrum. (*b*) Composite graph of the spectra of the scattered light for various laser detunings. The laser intensity is 174 mW cm^{-2} $\pm$ 50 mW cm^{-2}.

restricting the instrumental width of the Fabry–Perot to less than 20 MHz, thus allowing the sidebands on the emission spectrum to be well resolved. The Fabry–Perot, described by Hercher (1968), has a free spectral range of 750 MHz and is scanned in 5 seconds. The output of the photomultiplier following the Fabry–Perot drives the vertical input to an oscilloscope while the horizontal time base is set at 1 s cm^{-1}. Photographs of the traces are taken and the data is later reduced to the form shown in figures 2 and 3. A typical oscilloscope trace is also included in figure 3.

On the receding axes of figures 2 and 3 are plotted the laser detuning from the absorption line centre. The emission spectra for various detunings are plotted horizontally. The horizontal scale is centred on the laser frequency and not on the absorption line centre. We have observed that the central peak in every case coincides with the laser frequency to within less than 20 MHz. This conclusion was reached by simultaneously observing the spectra of both the scattered light and a portion of the incident laser beam.

The receding scale in figure 2(*b*) and 3(*b*) is calibrated by monitoring the scattered intensity. If the spectrally-integrated scattered intensity is plotted as a function of the frequency of the applied optical field one gets the power-broadened absorption spectrum. However, in this case, for the relatively high input intensities of 406 mW cm^{-2} and 174 mW cm^{-2}, the curve is asymmetric. This is due to another hyperfine transition at a frequency just 60 MHz lower than the $F = 2 \rightarrow 3$ transition (see figure 2(*a*)). This other transition is a three-level transition and at the 406 mW cm^{-2} power density these two transitions are mixed. The three-level transition provides a path for the escape of the atomic population from the two-level system by optical pumping. Thus, on the low frequency side of the $F = 2 \rightarrow 3$ transition, the scattering falls off much more rapidly with frequency than on the high frequency side. This effect is indicated in figure 2(*b*) by the skewed enveloped which fits the peaks of the spectra going along the receding axis.

The presence of this three-level system is further verified by noting in figure 2 and 3 the frequency component at +1770 MHz on the high frequency side of the spectrum when the laser is tuned below the resonance frequency. The laser partially excites the three-level transition and two frequencies are radiated, one at the laser frequency and another 1770 MHz away when an atom is optically pumped into the third level.

The spectra in the centre and lower right of figures 2 and 3 show quite clearly the sidebands generated by Stark splitting. As the laser intensity is increased, the sidebands are spread further from the central peak. In addition, one can see that as the detuning is increased, the sidebands spread out.

If we attempt to explain these results using phenomenological semiclassical equations (optical Bloch equations), we find that they predict that the atoms will suffer optical nutations for a few radiative lifetimes after they enter the interaction region and then they will settle down to the saturation limit with their dipole moments oscillating harmonically at the applied field frequency (Tang and Silverman 1966, Hocker and Tang 1968, Hoff *et al* 1970, Brewer and Shoemaker 1973). In our experiments each atom interacted with the field for approximately 100 radiative lifetimes. By using the slit of figure 1 we have determined that the spectrum of the light scattered by the front half of the illuminated region is identical to that from the second half. We do not observe a sharp line spectrum even for light scattered from atoms which have been in the laser beam for 50 lifetimes or more. Thus it would appear that the optical Bloch equations are inadequate for this problem. Because of the importance of spontaneous emission in this problem, some of the theoretical attempts to predict the spectrum have used quantum electrodynamics (Stroud 1973). Unfortunately, because of the difficulty of the

QED calculations, they have been carried out only for the initial transient regime; the long term solutions are unknown. There may or may not be a 'saturation steady-state limit' as in semiclassical theory (Brewer and Shoemaker 1973). The results of these calculations predict a spectrum qualitatively similar to that measured above but differing in some respects. In particular the predicted ratio of the heights of the central peak to the sidebands is 2:1 at resonance (Stroud 1973). Secondly, the areas under the sidebands are predicted to differ from each other appreciably for the detunings studied, whereas experimentally they appeared nearly equal when the laser was tuned to the high frequency side of the transition. These discrepancies are possibly due to our failure to include the nearby three-level transition in the theoretical calculations. We are carrying out a more detailed analysis and will describe our results shortly in an extended paper. One other theoretical complication has been investigated and found not to affect the predicted results. The two resonant levels are made up of 12 degenerate Zeeman sublevels. (We have experimentally cancelled the earth's field so that these levels are very accurately degenerate.) There are a number of slightly differing dipole matrix elements between various pairs of Zeeman sublevels, but they differ by too little to lead to resolvable details in our experiment.

Though the theoretical analysis of this experiment is not yet complete, we are able to reach two conclusions: (i) The spectrum of a resonantly scattered monochromatic field consists of a central peak at the applied field frequency and two symmetrically placed sidebands, and (ii) This spectrum persists when phenomenological semiclassical theory predicts that the dipole moment should have settled down to a harmonic oscillation.

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