Asymmetric Line Shapes for Weak Transitions in Strong Standing-Wave Fields

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(Received 31 December 1986)

We have observed the resonance line shape for a very weak atomic transition excited when an atomic beam intersects a strong standing-wave laser field. The line shape has a dramatic intensity-dependent distortion which is Doppler free and independent of the excitation rate. We have calculated the line shape predicted by optical Bloch equations that include a spatially varying ac Stark shift, and find good agreement with our experimental results.

PACS numbers: 32.70.Jz, 32.80.−t

An isolated atom which is weakly excited in a standing-wave field is one of the most basic systems one can study in spectroscopy. It has been commonly assumed that this simple case would have a symmetric, easily understood line shape. We have observed that this is not the case, and in fact very weak transitions in a strong standing-wave field can show striking asymmetric distortions of the resonance line shape. This is significant because these are the conditions encountered in the ongoing precision measurement of parity nonconservation in cesium,[1] and this measurement relies on a thorough understanding of the resonance line shape. Also this is relevant to precision wavelength standards for which weak transitions have been sought because of their narrow linewidths. We find that the intensity-dependent distortion has two rather interesting and surprising characteristics. First, the frequency dependence is characterized by the natural linewidth, and, although the distortion depends on the laser intensity, it is independent of the excitation rate. Thus we have the unusual situation that on a Doppler-broadened single-photon resonance line there is a distinct Doppler-free structure for arbitrarily small excitation rates. Second, the frequency dependence of the distortion for electric dipole ($E1$) transitions is the mirror image of that for magnetic dipole ($M1$) transitions. We have studied this distortion of the line shape for a variety of experimental conditions, and find our results agree very well with the line shape we calculate using optical Bloch equations that include the spatially varying ac Stark effect.

Prentiss and Ezekiel[2] have previously studied the intensity dependence of the line shape of the sodium D line. They observed an asymmetry at moderate to high excitation rates which they explained in terms of the induced dipole force changing the atomic trajectories. The asymmetry that we observe is different, though, in that it is independent of the excitation rate. To our knowledge this is the first observation of this type of line-shape distortion. In a rather different context, Roso *et al.*[3] have predicted spectral features due to a spatially inhomogeneous ac Stark shift. Their predictions concerned the nonlinear absorption on a probe transition which had a common level with a strongly driven transition.

For our study of the line shape, a beam of atomic cesium is excited in an intense standing-wave field. We observe the $6S$ to $7S$ transition, which is extraordinarily weak, and this is not power broadened even in very high fields. Before presenting the experimental results, let us discuss the $6S$-$7S$ transition in cesium, because it has some features that are different from normal transitions. A detailed discussion of this is given in Ref. 1; here we shall simply summarize the results.

As shown in Fig. 1, both the $6S$ and $7S$ states are split into two hyperfine states with total angular momenta of $F=3$ and 4. In the absence of an electric field there are four possible $M1$ transitions, with matrix elements that

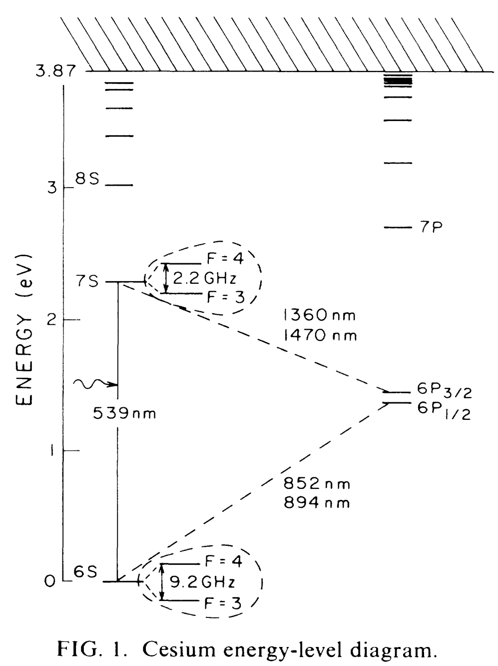

FIG. 1. Cesium energy-level diagram.

are a few times $10^{-5}\mu_B$. In the presence of a dc electric field, these transitions acquire Stark-induced $E1$ transition amplitudes given by

$$A_{St} = \alpha \mathbf{E}\cdot\boldsymbol{\epsilon}\delta_{FF'} + \beta|\mathbf{E}\times\boldsymbol{\epsilon}|, \tag{1}$$

where ϵ is the oscillating electric field, $\mathbf{E}$ is the static field, and α and β are the scalar and vector transition polarizabilities. Since $\alpha/\beta = 10$, the $\Delta F = 0$ transition rates can be 100 times larger than those of the $\Delta F = +1$ or -1 lines. By choice of a large enough $\mathbf{E}$ the Stark-induced $E1$ amplitude can be made much larger than the $M1$ amplitude. Although in general these two amplitudes can interfere, this does not happen when they are excited by a standing-wave field.

The basic experimental setup is shown in Fig. 2. Except for minor modifications, it is identical to that discussed in Ref. 1. The output of a continuous-wave dye laser goes into a resonant Fabry-Perot interferometer. This acts as a power-buildup cavity and provides an intense standing-wave field. A collimated beam of atomic cesium intersects this field at right angles, and a static electric field can be applied to the intersection region. The $6S$-$7S$ transition rate is monitored by observation of the 852- and 894-nm light emitted on the $6P$-$6S$ portion of the $7S$ cascade decay.

The dye laser produces up to 300 mW of light at 540 nm. The laser frequency is locked to the resonant frequency of the power-buildup cavity by electronic feedback, and has a residual jitter on the order of 25 kHz. The laser light is matched into the lowest-order mode of the power-buildup cavity. By use of a piezoelectric transducer the cavity length is adjusted to tune the resonant frequency over the atomic transition. The intracavity standing wave is composed of two traveling waves, each with 1600 times the power of the incident laser beam. This is about a factor of 10 higher power than that used in Ref. 1. The beam in the cavity has a Gaussian wave front with a diameter of 0.050 cm and a divergence of about 2×10^{-3} over the 2-cm length where it intersects the cesium beam. The cesium beam is produced by a large-area microchannel plate nozzle followed by a multiple-slit collimator. This collimation reduces the residual Doppler broadening of the $6S$-$7S$ transition to 24 MHz. By tilting the collimator, we are able to adjust the intersection angle between the atomic beam and the laser beam around its nominal value of 90 deg. The fluorescence signal is obtained by use of a cylindrical mirror to image the interaction region onto a silicon photodiode.

In Fig. 3(a) we show the line shape observed at relatively low intracavity power. The width is determined by the residual Doppler broadening. In Fig. 3(b) we show a

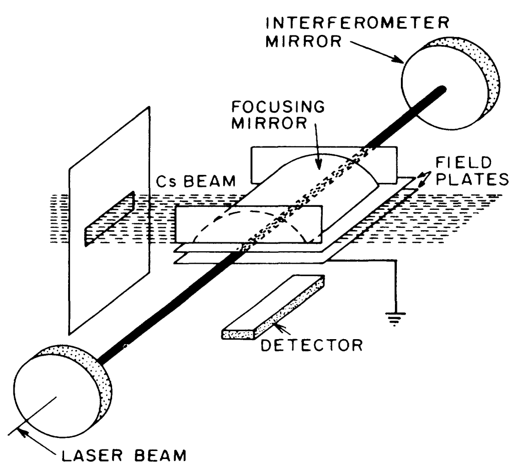

FIG. 2. Schematic of interaction region.

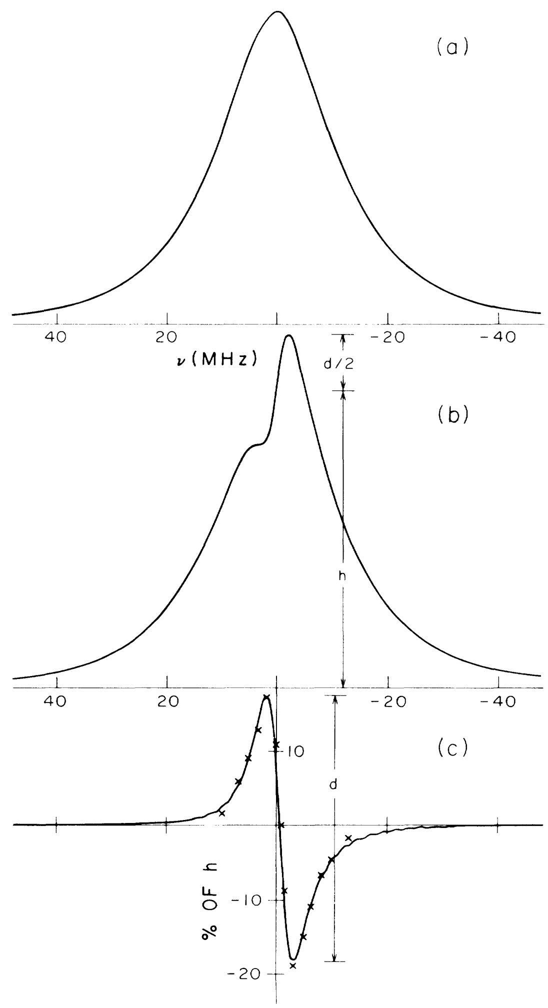

FIG. 3. (a) Line shape of $E1$ transition excited by 13.7 W of power in each of the two traveling waves in the standing wave. (b) Same transition with 277 W per traveling wave and a different vertical scale. (c) The curve corresponds to the curve in (a) times 277/13.7 minus the curve in (b). The crosses are the points given by the theoretical calculation.

high-power line shape and in Fig. 3(c) we show the difference that is obtained by our scaling the curve in Fig. 3(a) by the laser power ratio and subtracting it from the curve in Fig. 3(b). There is no absolute frequency scale, and so the origins of the two frequency scales were set so as to superimpose the undistorted wings of the lines. At moderate power (less than 100 W) the difference curve passes through zero at essentially the undistorted line center, and the width, taken to be the separation between the positive and negative peaks, is always 3.5(5) MHz. This matches the 3.3-MHz natural linewidth of the $7S$ state. As the power is increased above 100 W the curve broadens and shifts slightly, as can be seen in Fig. 3(c). For the highest power we used (350 W), a width of 5 MHz and a shift of about 0.5 MHz were observed. This power corresponds to a peak intensity of 3.5×10^5 W/cm^2 at the center of the Gaussian wave fronts of each of the traveling waves in the cavity.

In Fig. 4 we show the dependence of the distortion on the laser power. We are characterizing the asymmetry in terms of the fractional distortion, D, which we define as the ratio of the size of the distortion (d) to the peak height (h) the line would have if there were no distortion. This value initially rises linearly, but at higher power it begins to saturate.

The line shape for $E1$ transitions was found to be independent of laser polarization, static electric field, and hyperfine transition ($\Delta F=\pm1$ and 0). While the line shape remained the same as these conditions were changed, the transition rate per atom varied between 1 s^{-1} and 10^4 s^{-1}. When we observed an $M1$ transition by setting the dc electric field to zero, the line shape was the mirror image of that of the $E1$ transition. The rate was enhanced on the high-frequency side and suppressed on the low-frequency side of the line. We also found

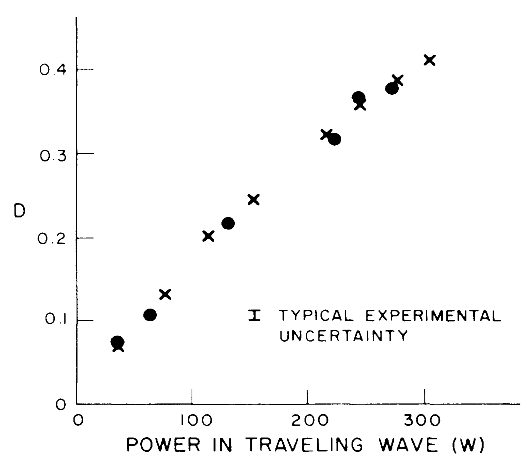

FIG. 4. The fractional distortion D vs traveling-wave power. The crosses are the theoretical results and the dots are experimental points.

that when the angle between the cesium beam and the atomic beam differed from perpendicular by a value greater than the divergence of the cesium beam, the line shape began to split into a doublet corresponding to excitation by each of the Doppler-shifted traveling waves, and the intensity-dependent distortion went away.

Although the explanation is somewhat subtle, these results can be accounted for by consideration of how the ac Stark shift in a standing wave affects the line shape. This Stark shift arises because the field couples both the $6S$ and $7S$ states with the P states of the atom. Since the coupling to all of the P states is far off resonance, the shift is essentially constant over our frequency range. Also it is independent of the polarization of the laser. Considering only the coupling to the P states with principal quantum numbers 6, 7, and 8, we calculate the ac Stark shift of the transition frequency to be 6.8×10^{-3} Hz(V/cm)$^{-2}\epsilon^2$, or 21 Hz cm^2/W times the intensity in one traveling wave. There is an uncertainty in this result due to the contribution from higher states and errors in the calculated matrix elements, but we estimate that the result is accurate to within a few percent. This shift was incorporated into the usual optical Bloch equations for a two-level system by our simply replacing the energy difference between the states, E_1-E_2, by the Stark shifted difference, $E_1-E_2+\delta(t)$. The ac Stark shift, $\delta(t)$, is proportional to $\epsilon^2\cos[kz(t)]$, where $z(t)=z_0+v_zt$ with z_0 being an initial position and v_z representing an atomic velocity transverse to the laser wave fronts. Also ϵ still contains the Gaussian x and y dependence of the wave front. We can neglect transitions to other states in the system because the photoionization rate is much smaller than the $7S$-$6P$ decay rate.[4]

We have solved the Bloch equations numerically to find the time-integrated population in the $7S$ state for an atom in the laser beam, as a function of laser frequency. The observed fluorescence is proportional to this population for constant velocity across the beam. The solution involves averaging over the distribution of transverse velocities and initial positions z_0 in the standing wave, while taking into account the Gaussian field distribution of $\epsilon(x,y)$ in the x-y plane. To simplify the solution, we assume that $\epsilon(x,y)$ is constant over distances in the x-y plane corresponding to that moved by an atom in an atomic lifetime, and that the atoms follow straight-line trajectories. These assumptions are reasonably good since a typical atom takes more than 20 lifetimes to cross the laser beam and is only deflected by about 10^{-3} rad by the dipole force. The calculated line shapes, particularly the intensity-dependent distortions, agree very well with all our observations. Examples of this can be seen in Figs. 3(c) and 4, which have been calculated with *no* free parameters. The basic shape of the distortion matches surprisingly well with what we observe and the height matches to within the uncertainty of the calculated ac Stark effect. Even such subtle features as the slight power-dependent shift in the frequency at which the difference curve crosses through zero and the small difference in height between the positive and negative peaks are reproduced in the calculated line shapes.

We can understand these results qualitatively by considering the relationship between the Doppler shift of an atom and the ac Stark shift it will have. If an atom has a large v_z it will move rapidly across the wave fronts of the standing wave. In one lifetime it will sample and thus average over many periods of the field, and hence of the ac Stark shift. The perturbed resonant frequency of this atom will then be the unperturbed energy difference shifted by the spatial average of the Stark shift. The spectrum for a group of such atoms will be two Lorentzian peaks which have the natural linewidth and are Doppler-shifted symmetrically above and below the perturbed resonant frequency. All large velocities show similar behaviors; thus the wings of the line, which are due to these velocities, will be symmetric, and the shape of the wings, though not the central frequency, will be independent of the intensity. For very low-velocity atoms the spectrum is quite different, however. A slow atom's position is nearly constant during one lifetime $[z(t) \approx z_0]$ so that its resonant frequency will be shifted by the $\delta(t)$ determined by the field at that particular point in space. The spectrum one obtains for a spatial average of such atoms is a single peak skewed toward the maximum value for $\delta(t)$, because the high-field regions have the largest transition rate as well as the largest ac Stark shifts. The combination of these two very different spectra for high- and low-velocity atoms results in the line shape we observe. The velocity characterizing the distinction between high- and low-velocity regimes is that at which an atom goes one wavelength in a lifetime. At this velocity an atom will have a Doppler shift equal to the natural linewidth; this is why the characteristic frequency scale is the natural linewidth.

From this picture it is also quite easy to understand the frequency dependence of the $M1$ transitions. In a standing wave the antinodes of the oscillating magnetic field occur at the position of the electric-field nodes. This means that the $M1$ transitions for low-velocity atoms will occur primarily in regions of low electric field and hence be skewed toward small ac Stark shifts. Since this is just the opposite of the $E1$ case, the distortion of the line shape is reversed.

In conclusion, we have found that the simple case of a weak transition excited in a standing-wave field does not have a symmetric line shape. The line shape has an intensity-dependent asymmetry with characteristics that are unlike previously observed distortions of spectral lines. However, these characteristics can be fully explained if one carefully considers the combined effects of the Doppler and ac Stark effects.

We are happy to acknowledge useful discussions with Dr. S. Gilbert, Dr. M. Prentiss, Dr. C. Tanner, and assistance in the computations from Dr. D. Kelleher. This work was supported by the National Science Foundation and one of us (C.E.W.) received support from the Alfred P. Sloan Foundation.

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