Observation of Intensity-Dependent Fluorescence Line-Shape Asymmetry for Two-Level Atoms in a Standing-Wave Field

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Intensity-dependent fluorescence line-shape asymmetries have been observed for two-level atoms in a standing-wave excitation field. These asymmetries are attributed to the force experienced by the induced atomic dipoles in the field gradient of the standing wave. Preliminary calculations support the experimental observations.

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We have observed intensity-dependent asymmetries in the fluorescence line shape of a two-level sodium atomic beam in the presence of either a traveling- or a standing-wave excitation field. The asymmetry in the traveling-wave case has been noted previously and is due predominantly to the recoil that the atoms experience in traversing the excitation field.[1] We attribute the observed asymmetries in the standing-wave field to the force experienced by the induced atomic dipoles in the field gradient of the standing wave.

Such line-shape studies are of much interest because of the important information that they convey about basic atom-field interactions and also because such line-shape asymmetries can influence the accuracy of frequency and wavelength standards as well as the ultimate precision of spectroscopic measurements.

Our experimental setup, shown in Fig. 1, consists of a sodium atomic beam which interacts at right angles with two collimated beams from two dye lasers. Both laser beams are circularly polarized (σ^+) to better than 99%.

The first laser beam, i.e., the state-preparation (SP) field, which is resonant with the $3^2S_{1/2}(F=2)$ to $3^2P_{3/2}(F=3)$ transition at 5890 Å, optically pumps the atoms in the $3^2S_{1/2}(F=2)$ ground state into the $m_F=2$ sublevel. The fluorescence induced by this beam is monitored by photomultiplier (PMT) No. 2 and the frequency of the SP field is held at the center of the resonance by a servo loop.

The atoms then interact with the second laser beam, i.e., the excitation field. Since the excitation field is σ^+ polarized, the only electric-dipole-allowed transition from the $3^2S_{1/2}(F=2,m_F=2)$ sublevel is to the $3^2P_{3/2}(F=3,m_F=3)$ sublevel, so that the atoms may be considered a two-level system.[2] The fluorescence induced by this field is monitored by PMT No. 1.

Figure 2 shows typical fluorescence line shapes for two excitation field intensities, where the $1/e^2$ radius of the field was 1.5 mm. Figures 2(a) and 2(c) correspond to a traveling-wave excitation and Figs. 2(b) and 2(d) to a standing-wave excitation. The standing wave was generated by reflection of the excitation field back on itself using a cat's eye reflector. The reflected intensity was greater than 97% of the incoming intensity. An acousto-optic frequency shifter was inserted in front of the laser to prevent optical feedback.

The weak-field line shapes for peak intensity $I_0=0.1$ mW/cm^2 are clearly symmetric in either a traveling-

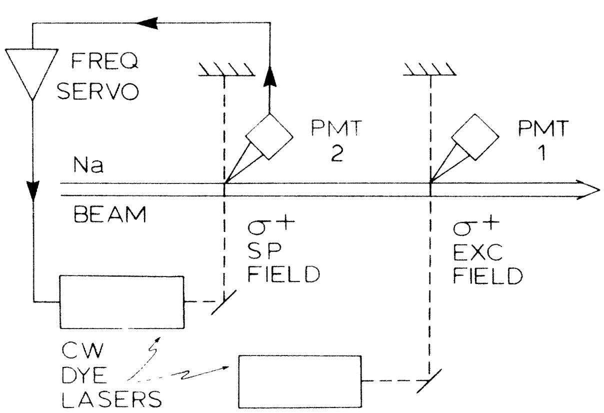

FIG. 1. Experimental setup.

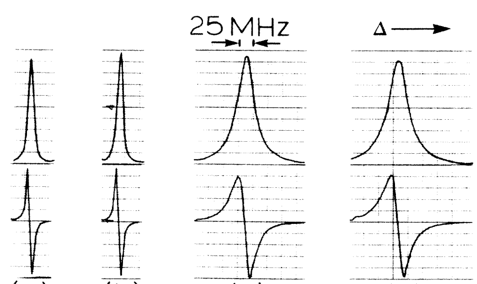

FIG. 2. Fluorescence line shapes for the $3^2S_{1/2}(F=2)$ to $3^2P_{3/2}(F=3)$ transition. (a),(b) Weak traveling-wave and standing-wave excitation, respectively ($I_0=0.1$ mW/cm^2); upper traces are fluorescence line shapes [vertical scale on (b) is 0.56× (a)] and lower traces correspond to the derivatives of the line shapes [vertical scale of (b) is 0.56× (a)]. (c),(d) Corresponding line shapes for $I_0=50.0$ mW/cm^2; upper traces have the same vertical scale, and the vertical scale on the lower trace in (d) is 1.26× (c).

wave [Fig. 2(a)] or a standing-wave field [Fig. 2(b)]. At higher intensities ($I_0 = 50.0$ mW/cm^2) [Figs. 2(c) and 2(d)] the traveling-wave and standing-wave line shapes become asymmetric.

One way to quantify the observed asymmetry is to consider the derivative of the line shape. The peak height of the derivative is a measure of the maximum slope of the line; therefore, a measure of the line-shape asymmetry is given by ϵ, the magnitude of the ratio of the peak height of the derivative for a detuning $\Delta < 0$ to the peak for $\Delta > 0$. If the line were perfectly symmetric, ϵ would be unity.

Derivativelike line shapes were generated by modulation of the excitation-laser frequency at a rate f_{m1}, and synchronous demodulation of the detected fluorescence. The modulation excursion was 400 kHz peak to peak which is much smaller than the 10-MHz natural linewidth of the transition. Figure 2 shows such derivativelike line shapes below the corresponding fluorescence line shapes.

The general behavior of ϵ as a function of intensity is shown in Fig. 3 for a standing-wave and also a traveling-wave excitation field. At very low intensities ($I_0 < 0.2$ mW/cm^2) the line shapes are symmetric for either excitation field, as indicated in Fig. 2. For I_0 between approximately 0.2 and 5.0 mW/cm^2 both line shapes become asymmetric, but the direction of the asymmetries are opposite. At intensities above 5.0 mW/cm^2 the asymmetry in the fluorescence line shape for a standing-wave excitation field changes sign. At still higher intensities ($I_0 = 30$ mW/cm^2), the asymmetry in the traveling-wave case begins to decrease, but the standing-wave asymmetry continues to increase.

In addition to measuring the asymmetries in the line shapes, we measured the frequency shift, δ, of the fluorescence maximum as a function of intensity. In order to measure this shift we used the same laser as a source for both the state-preparation beam and the excitation field. A frequency shifter was inserted in the SP field so that the frequencies of the two beams could be controlled independently. We determined δ by adjustment of the frequency shifter in the SP field until the demodulated fluorescence induced by the SP field and that induced by the excitation field were both zero.

Figure 4 is a plot of δ as a function of intensity. The plot shows that for a traveling-wave excitation, δ is positive (i.e., the peak fluorescence is shifted toward higher frequencies), and that for a standing-wave excitation, δ is negative regardless of the direction of the accompanying line-shape asymmetry.

The shift in the frequency of the fluorescence maximum and the accompanying line-shape asymmetry in the traveling-wave case are, as was mentioned earlier, due to atomic recoil.[1] We performed numerical calculations of the recoil-induced asymmetry (the dotted curve in Fig. 3) for the traveling-wave excitation in our experiment and the results are in good agreement with the experimental data. Velocity-diffusion effects due to spontaneous emission may also be important in some configurations, but were not significant in this

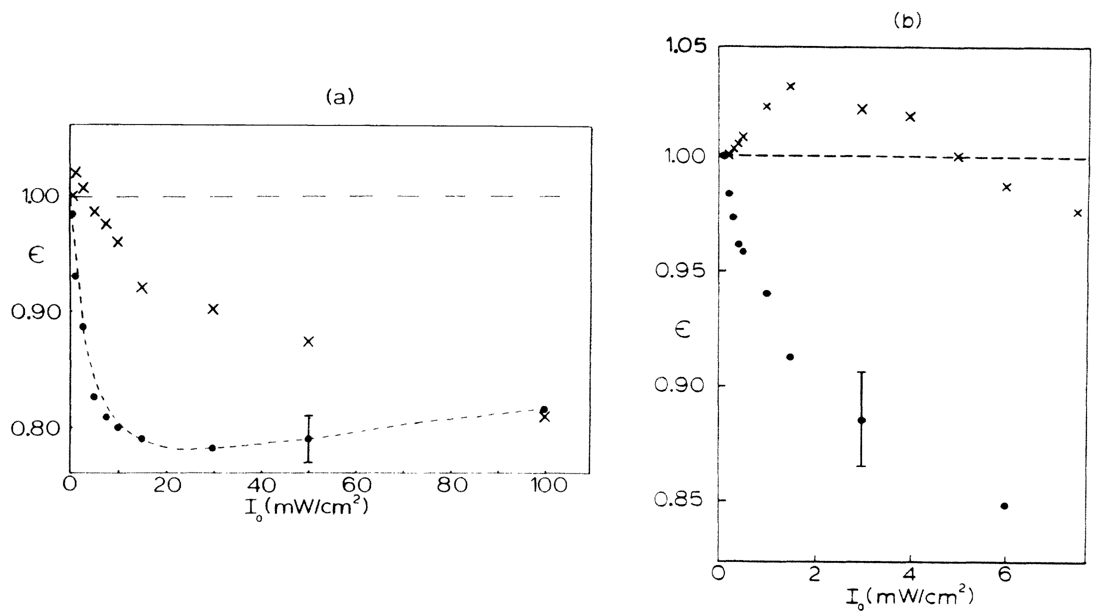

FIG. 3. Observed fluorescence line-shape asymmetry, ϵ, as a function of intensity for a traveling-wave (dots) or a standing-wave (crosses) excitation field. The dotted curve is the calculated line-shape asymmetry for a traveling-wave excitation. Part (b) is an expanded-scale view of the low-intensity region of (a).

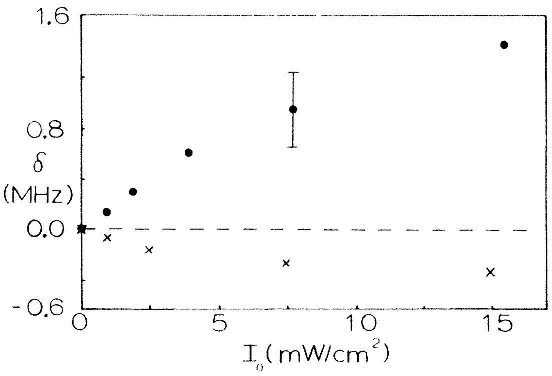

FIG. 4. Detuning of the fluorescence maximum, δ, as a function of intensity for a traveling-wave (dots) or a standing-wave (crosses) excitation field.

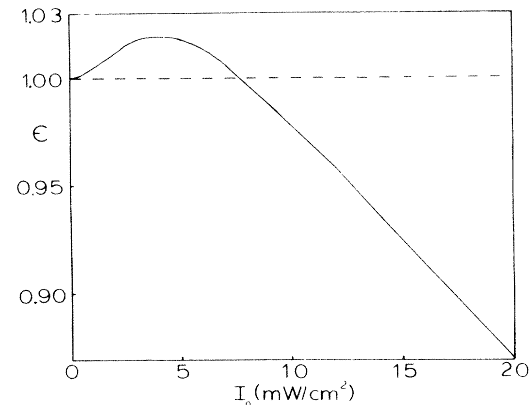

FIG. 5. Preliminary calculation of the fluorescence line-shape asymmetry for a standing-wave excitation field.

case because the power-broadened linewidth increased more rapidly than the Doppler broadening due to velocity diffusion.

We will now consider the case of a standing-wave excitation. If an atom has no velocity component along the direction of the standing wave, then the atom is equally likely to absorb a photon from either of the two traveling waves that compose the standing wave, so that there will be no net velocity transfer due to atomic recoil, but only a diffusion of the atomic velocities caused by both spontaneous and stimulated emission as will be discussed later. This diffusion of the atomic velocities will result in a Doppler broadening of the line, but will not by itself induce an asymmetry.

We believe that the asymmetry that we observe in the standing-wave case is the result of atomic motion due to the force on the induced atomic dipole in the field gradient of the standing wave. This force pushes atoms toward the high-intensity regions when the excitation field is detuned below resonance and toward the zero-intensity regions for an excitation field detuned above resonance[3]; therefore, for short interaction times and $\Delta < 0$, the atoms congregate in the high-intensity regions and so the total fluorescence will be higher than that for a uniform distribution. Similarly, for $\Delta > 0$ the fluorescence will be lower than that for a uniform distribution. Thus this frequency-dependent nonuniformity in the atomic density causes a distortion of the fluorescence line shape, and a shift in the frequency of the fluorescence maximum toward lower frequencies.

To verify this interpretation of the asymmetry we performed a preliminary calculation that considered a uniform atomic beam with a single velocity in a direction perpendicular to the standing-wave field and an observation region of approximately 0.1 mm^2. A plot of the result of this calculation is shown in Fig. 5. For intensities up to approximately 20 mW/cm^2 the calculated asymmetry is in reasonable agreement with the data, but at higher intensities the asymmetry predicted by this simple model is larger than that observed in the experiment.

To explain the behavior at higher intensities it may be necessary to include the effects of other processes besides the dipole force. These include the diffusion of the atomic velocities due to spontaneous and induced emissions as well as effects due to velocity components along the standing wave.

There are two separate velocity-diffusion mechanisms for a standing-wave excitation field: one associated with spontaneous emission and one associated with the stimulated processes.[4] The spontaneous velocity-diffusion effect results from the randomness in the direction of the recoil due to spontaneous emission. The stimulated diffusion effect is more complicated, but can be viewed as the result of fluctuations in the dipole force due to the interaction between the field gradient and the fluctuating induced atomic dipole moment.[5] These diffusive effects will introduce a spread in the atomic velocities which will reduce the density-nonuniformity effects of the dipole force and will thus reduce the line-shape asymmetry, particularly at high intensities.

We will now consider the effect of a velocity component along the standing wave. Such a velocity component can arise either from a finite initial divergence in the atomic beam or as a result of the interaction between the atomic beam and the excitation field. For atoms with a velocity component along the standing wave, one traveling wave will appear shifted to the blue and the other traveling wave will appear shifted to the red; therefore, the expressions for the dipole force and fluorescence will be different from those for a stationary atom.[6] Furthermore, the probability of absorption of a photon from the traveling wave closer to resonance is greater than that for the other traveling wave, so that the atom experiences a net recoil in the direction of the traveling wave that is closer to resonance. If $\Delta < 0$, this effect will decrease the component of the atomic velocity in the direction of the standing wave, so that motion along the standing wave will be damped; however, for $\Delta > 0$, the force will increase the velocity component in the direction of the standing wave so that motion along the standing wave will be enhanced.[7]

To test the conjectures made above, we have performed additional calculations which included the effects of atomic diffusion and a nonzero velocity along the standing wave on the fluorescence line-shape asymmetry. The results, which are still preliminary, predict that the line-shape asymmetry is reduced particularly at high intensities. In addition, we have conducted experiments that show that increasing the velocity along the standing wave decreases the observed asymmetry.

Finally, in order to fit the experimental data, the calculations must also include our experimental conditions, i.e., the angular spread in the atomic beam, the Gaussian distribution of the laser field, and the velocity distribution in the direction of the atomic beam.

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