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Determination of the branching of the $^1P_1$ Ba I level from studies of the intensity dependence of resonance fluorescence

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The resonance fluorescence from the ^{138}Ba I 5535-Å $6s^2\,{}^1S_0$–$6s6p\,{}^1P_1$ transition has been measured as a function of incident intensity using a Doppler-free atomic beam geometry and a single-mode cw dye laser. The upper $^1P_1$ level has a weak branch to intermediate metastable D states. A long laser-atom interaction region was used, which ensured that essentially all atoms could be pumped into the D states during their transit through the laser beam. By analyzing the shape of the saturation curve a value for the branching ratio to the $6s5d\,{}^{1,3}D$ levels of $(3.57\pm0.38)\times10^{-3}$ has been obtained.

I. INTRODUCTION

Atoms or ions with excited states which decay almost exclusively back to the ground state approximate two-level systems when illuminated with laser light near resonance. Such "recyclable" transitions have been used in many types of experiments such as isotope separation via photodeflection,[1] cooling and trapping[2,3] of atoms and ions, optical pumping for nuclear orientation,[4] and high sensitivity laser spectroscopy.[5,6] For most atoms and ions, experiments of this nature are limited by branching from the upper electronic level to other intermediate, usually metastable, states.

In the $6s^2\,{}^1S_0$–$6s6p\,{}^1P_1$ 5535-Å resonance line of atomic barium, the upper $^1P_1$ state branches only weakly to the metastable $6s5d\,{}^1D$ and ^3D states. The relevant energy levels are shown in Fig. 1. In several of the laser spectroscopic studies mentioned above[1–6] this branching has been a significant limitation of the measurements. In recent measurements[7] of the time dependence of the intensity of light resonantly scattered by individual barium atoms, which provided experimental evidence for quantum jumps, a knowledge of the strength of the $^1P_1$-$^{1,3}D$ transition played a major role in the interpretation of the data.

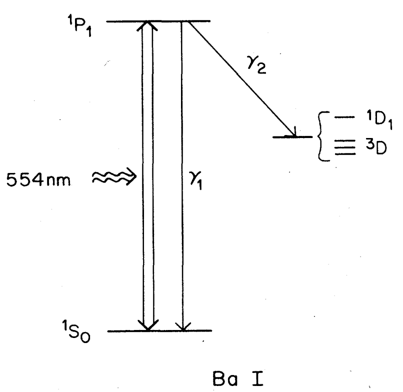

FIG. 1. Relevant energy levels in the Ba I spectrum. The D-state lifetimes are $\simeq0.5$ sec (Ref. 13) and transitions back to the ground state play no role in the present experiments.

The two most recently published values of the summed strengths for branching from the barium $6s6p\,{}^1P_1$ level to the $6s5d\,{}^1D$ and ^3D states were made using photodeflection[8] and by measuring the decay rate of the fluorescence from optically pumped barium atoms in a buffer gas.[9] The results from these determinations differ by more than an order of magnitude and underscore the difficulties involved in measuring the strengths of weak atomic transitions.[10]

This paper presents a determination of the summed strengths from the $^1P_1$ level to the D levels using a new method which should be relatively free from systematic errors. In the experiment the intensity of light resonantly scattered by ^{138}Ba atoms in a collisionless atomic beam was measured as a function of incident light intensity. Laser light perpendicularly intersected the atomic beam over a distance sufficiently long to ensure that almost all of the atoms were driven into metastable states at moderate laser intensity. This results in the scattered light saturating at lower intensity than for a pure two-level system, and an analysis of the saturation curve yields a value for the branching ratio.[11] The result is in agreement with a reanalysis[12] of the data of Ref. 9 and with the theoretical value of McCavert and Trefftz.[13]

II. EXPERIMENT

A diagram of the atomic-beam chamber is shown in Fig. 2. A thermal atomic beam produced by an oven, at temperature 467 °C and loaded with natural barium metal, was tightly collimated to have a transverse size of about $\frac{1}{2}$ mm in the interaction region where it was illuminated

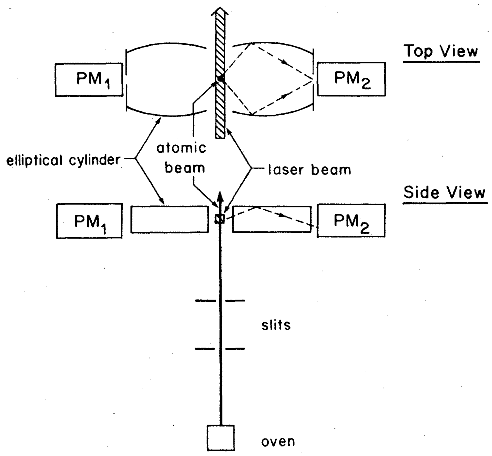

FIG. 2. Diagram of the atomic-beam chamber and light collector detector. Two orthogonal views are shown. The light collection efficiency is uniform over the laser—atomic-beam interaction region at the common line focus of the double elliptical cylinder.

with laser light. Approximately 10^5 atoms per second passed through the interaction region. The laser light was perpendicular to the direction of travel of the atoms and the collimation was such that it produced a negligible Doppler broadening of the resonance line [full width at half maximum (FWHM) 19 MHz (Ref. 14)].

The laser—atomic-beam interaction region was located at the common line focus of a polished double elliptical cylinder which served as a light collector. As shown in Fig. 2, resonantly scattered photons from the interaction region passed through slits at the opposite foci of the ellipses and onto two cooled photomultiplier tubes (EMI 9658). The top and bottom ends of the cylinder were capped with flat, highly reflective plates which constrained scattered photons to leave the light collector through the photomultiplier tube slits. For the measurements presented here, the atomic beam was illuminated over a longitudinal distance of about 5 mm. Over the size ($5\times\frac{1}{2}\times\frac{1}{2}$ mm^3 diameter) of the effective source emitting photons, the light collector efficiency is uniform to within 0.5% and the overall detection efficiency was measured[15] to be 0.017.

Light from the dye laser was spatially filtered, expanded, and truncated with rectangular slits in order to make the intensity spatially uniform. Light emerging from these slits was focused onto the interaction region using a 4-m spherical lens to illuminate a 5-mm portion of the atomic beam. A series of adjustable apertures inside the chamber minimized the background due to slit-scattered laser light. This scheme did not produce a beam with perfectly uniform intensity, and the actual intensity profile experienced by atoms was measured in $\frac{1}{2}$-mm steps using a photodiode masked with a pinhole. The response of the photodiode was calibrated with respect to a Scientech 36-0001 calorimeter for absolute intensity measurements.

A commercial tunable dye laser (Spectra Physics 380A) was used with its frequency locked to two external étalons giving a linewidth less than 1 MHz. The étalons were ultimately referenced to a transverse-Zeeman stabilized He-Ne laser (Laboratory for Science 220). It was possible to tune the dye laser frequency over a barium resonance peak with a long-term stability better than 0.3 MHz by varying the He-Ne frequency.[15]

The dye-laser frequency was centered on the ^{138}Ba resonance at low laser power to avoid Doppler shifts due to radiation pressure (discussed below). Both the laser frequency and the laser power were servo-locked to fixed values and the intensity at the interaction region was varied using a commercial attenuator (Newport Corporation 925-B). A large-area calibrated photodiode used in conjunction with a beam pickoff monitored the laser power, and the rates of detected photons were recorded using standard high-speed photon counting electronics. Small (≤ 0.016) deadtime corrections were applied to the high rate data.

III. ANALYSIS AND DISCUSSION

The variation of the on-resonance scattering rate with incident light intensity (the saturation curve) has been thoroughly studied for two-level atoms.[16] For illumination times greater than a few atomic lifetimes, as in the present case, the scattering rate is given by[17]

$$\frac{dn}{dt}=\frac{\gamma_1}{2}\frac{I/I_s}{1+I/I_s}\ , \tag{1}$$

where $I_s=\pi hc\gamma_1/3\lambda^3$, γ_1 is the decay rate to the initial state and λ is the wavelength. For an atom of speed v entering the beam at $z=0$, Eq. (1) becomes

$$\frac{dn(z)}{dz}=\frac{\gamma_1}{2v}\frac{I(z)/I_s}{1+I(z)/I_s}\ . \tag{2}$$

In the case of a three-level system with a decay rate γ_2 to a metastable intermediate state, the atoms can be divided into two classes: (1) atoms which continue fluorescing during their transit through the laser beam (the "transit-time"-limited case), and (2) atoms which branch to the intermediate state while in the laser beam (the "optical-pumping"-limited case).[18]

Transit-time-limited atoms behave as two-level systems during the measurement period.[7] Numerical integration of Eq. (2) over the laser spot using the measured variation of $I(z)$ yields the average number $\langle n_v\rangle$ of photons emitted by atoms of speed v during their transit.

Optical-pumping-limited atoms yield an average of γ_1/γ_2 photons. This average is the reciprocal of the branching ratio $\eta=\gamma_2/\gamma_1$.

Using the rate equations the average number of scattered photons for all atoms of speed v can be shown to be

$$\langle N_v\rangle=\frac{1}{\eta}[1-\exp(-\eta\langle n_v\rangle)]\ .$$

This expression is then averaged over the speed distribution of the atoms to yield the average number $\langle N\rangle$ of scattered photons for all atoms. Evaluation of $\langle N\rangle$ for various values of the laser power yields the saturation curve.

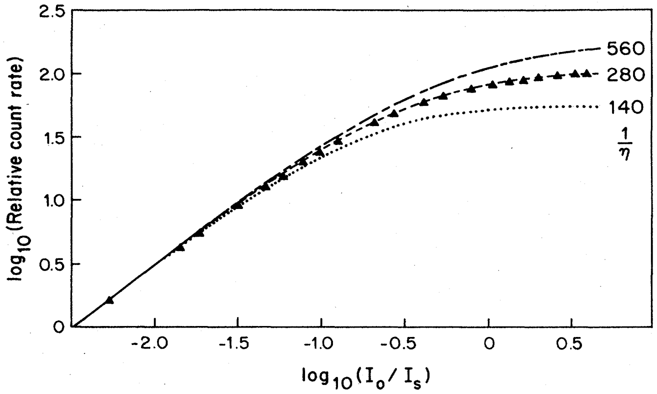

FIG. 3. Measured and calculated saturation curves with a long interaction region for the 5535-Å barium resonance. Reciprocal branching ratios of 140, 280, and 560 were assumed for the calculated curves and the saturation intensity I_s is 147 μW/mm^2.

When long interaction times are involved, as in the present work, atoms can be "pushed" partially off-resonance in their transit because of radiation pressure. This introduces an additional term into the denominator of Eq. (2) which becomes[19]

$$\frac{dn(z)}{dz} = \frac{\gamma_1}{2v}\,\frac{I(z)/I_s}{1+\dfrac{I(z)}{I_s}+4\left[\dfrac{2\pi h}{M\lambda^2\gamma_1}n(z)\right]^2}\,, \qquad (3)$$

where M is the mass of the atom. This form of the equation must then be used to obtain $\langle n_v \rangle$ for the transit-time-limited case.

The calculated saturation curves, including radiation pressure effects, and the experimental points are plotted in Fig. 3. The ordinate is the logarithm of the resonantly scattered photon rate, normalized to 2.0 at the highest point. The abscissa is the logarithm of the ratio of the incident laser light intensity in the central region of the beam (I_0) to the saturation intensity parameter I_s. Using the measured value[14] for γ_1, of 119×10^6 sec^{-1} yields $I_s = 147$ μW/mm^2. The calculated curves are normalized to the data at the lowest measured power. The best-fit curve used a value of $1/\eta = 280$. Curves using $1/\eta$ values of 140 and 560 are included in Fig. 3 for comparison.

The statistical errors in the data points are negligible. Relative errors (due, e.g., to fluctuations in laser power) are also small. Assuming a 1% error in the scattering rate, the minimum χ^2 per degree of freedom is 1.8 for a value of $1/\eta$ of 280. The largest systematic error in this value comes from the uncertainty in the absolute laser power. Consideration of systematic and fitting errors leads to a final value of 280 ± 30 for $1/\eta$.

If the radiation pressure effect is neglected [using Eq. (2) rather than Eq. (3) in the analysis] the fit to the data is worsened somewhat ($\chi^2 = 2.2$) but the value for $1/\eta$ is changed by less than 2%. In the present case, the natural linewidth was 19 MHz and the frequency shift due to radiation pressure was about 5 MHz for optically pumped atoms.

TABLE I. Summed decay rates for the $6s6p\,^1P_1-6s5d\,^{1,3}D$ Ba I transitions.

Reference	Summed rates (10^4 s^{-1})	$1/\eta$
Theory		
McCavert and Trefftz[a]	35	340
Hafner and Schwarz[b]	16.7	712
Experiment		
Miles and Wiese[c]	490	24
Bernhardt *et al.*[d]	(<17)	(>700)
Niggli and Huber[e]	21	550^{+100}_{-20}
Myers *et al.*[f]	(210 ± 40)	(57 ± 10)
Kallenbach and Kock[g]	$\simeq42$	$\simeq285$
Trajmar *et al.*[h]	39.7 ± 6.6	300 ± 50
This work	42.5 ± 4.0	280 ± 30

[a]Reference 13.
[b]Reference 21.
[c]Reference 22.
[d]Reference 8.
[e]Reference 23. This is a reanalysis of the data of Ref. 8.
[f]Reference 9.
[g]Reference 12. This is a reanalysis of the data of Ref. 9.
[h]Reference 20.

The present branching ratio is compared with previous theoretical and experimental values in Table I. It is in agreement with the result of Ref. 12 which is a correction to the value given in Ref. 9, and also with the results of McCavert and Trefftz[13] and Trajmar.[20]

In summary, a new method which should be relatively free from systematic error has been used to determine the summed strengths from the $6s6p\,^1P_1$ to the $6s5d\,^{1,3}D$ states in atomic barium. It has the advantage that no levels are populated other than those involved in the branching process. The experimental parameters are well understood and well determined, and the analysis is straightforward. The method has limited applicability, but is well suited to intrinsically interesting energy-level configurations which approximate two-level systems with weak branching out of one of the two levels.

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