

Optical pumping to the $6s5d$ states in atomic barium

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Using a flow apparatus, the time dependence of $6s^2S_0-6s6p^1P_1$ (553.5 nm) resonance fluorescence in atomic barium has been studied following switch-on of cw laser excitation. It was found that the fluorescence decays with rates as great as $1.3 \times 10^6 \text{ s}^{-1}$. Assuming this can be ascribed to the pumping of atoms to the $6s5d^1D$ and 3D metastable levels, a value for the sum of the transition probabilities can be extracted.

INTRODUCTION

Recently there have been several studies involving the optical excitation of the $6s^2S_0-6s6p^1P_1$ resonance line of atomic barium, which are sensitive to the weak decay branches of the $6s6p^1P_1$ level to the $6s5d^1D$ and $6s5d^3D$ metastable levels (see Fig. 1). Among these are investigations of the effects of intense pulsed excitation of high-density vapors,^{1,2} optical beam deflection for isotope separation,³ and optical pumping, with circularly polarized light, for nuclear orientation.^{4,5} In the last case, pumping of the atoms to the $6s5d$ levels limits the degree of nuclear orientation that can be achieved. In view of the discrepancies among existing values for the spontaneous decay rates of the $6s6p^1P_1$ level to the $6s5d^{1,3}D$ levels, we attempted to

monitor the optical pumping to the $5d^{1,3}D$ levels by determining the time dependence of the resonance fluorescence following switch-on of cw excitation.

EXPERIMENT

A schematic of the flow cell used in the experiment is shown in Fig. 2. Argon gas flowed through a resistively heated brass tube of 4-mm inner diameter containing a natural barium sample. The barium-seeded argon emerging from the tube then passed through the expanded light beam from a single mode ring dye laser tuned to the center of the 553.5-nm resonance line. Fluorescence from a well defined region centered on the intersection of the laser beam and argon stream was imaged onto the photocathode of a photomultiplier fitted with a narrow-band interference filter for 553.5 nm. Extensive baffling of the laser beam entrance and exit arms of the chamber reduced the scattered light reaching the detector to levels comparable to the Rayleigh

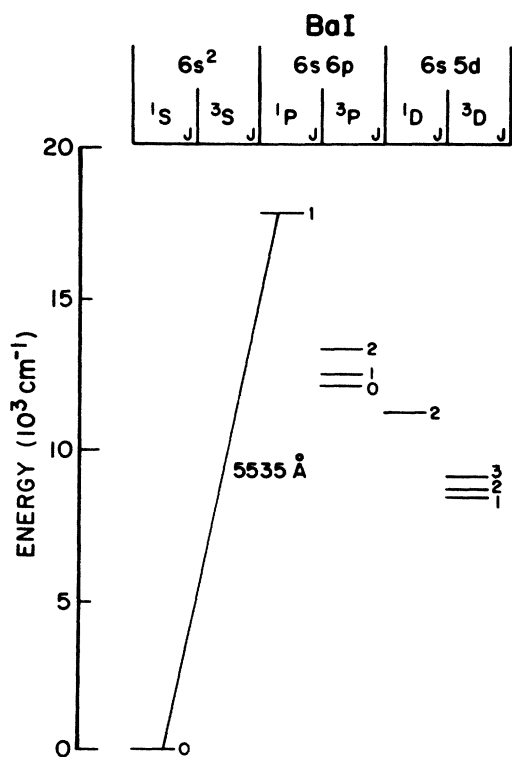


FIG. 1. Low-lying levels of atomic barium.

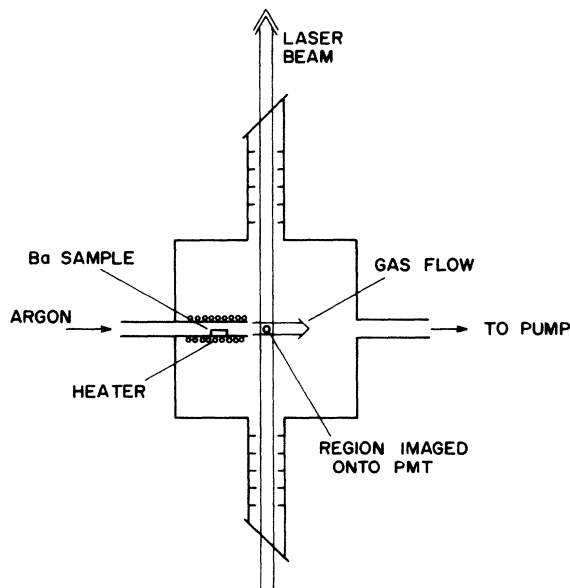


FIG. 2. Schematic of flow apparatus used to observe fluorescence.

scattering from the buffer gas (typically 100 torr argon). Consequently, good signal to noise was obtained with low-density, optically thin barium vapors.

The cw laser beam was temporally chopped using a pair of linear polarizers and a Pockels cell, to yield $\sim 20\text{-}\mu\text{s}$ -long, flat topped pulses ($\Delta I/I < 10\%$) with a rise time of about $1\ \mu\text{s}$. The laser intensity during the on period was typically $200\ \text{mW}/\text{cm}^2$. The signal from the phototube was fed into a boxcar analyzer to give the time dependence of the 553.5-nm fluorescence following switch-on of the laser pulse.

RESULTS AND ANALYSIS

The time dependence of the fluorescence was studied for a range of laser intensities, gas pressures, and laser spot sizes, and typical data are shown in Fig. 3. For low laser intensities ($\sim \mu\text{W}/\text{cm}^2$) the fluorescence was found to follow the laser intensity closely, while at higher laser intensities the fluorescence rose rapidly ($< 1\ \mu\text{s}$) to a maximum and then fell off exponentially with a time constant of several μs , to a steady-state value which depended on gas pressure and laser spot size.

The interpretation of this effect is that when the laser beam is switched on, fluorescence is observed from all the barium in the observation region, due to atoms cycling between the $6s^2\ ^1S_0$ and $6s6p\ ^1P_1$ states. But as a result of the spontaneous decay branch to the $6s5d\ ^1D$ and 3D metastable states, atoms gradually accumulate in these states, and fewer are available to be excited. The steady-state fluorescence results from the diffusion and flow into the observation region of unpumped ground-state barium atoms. The steady-state fluorescence relative to the peak was observed to increase as the laser beam spot size was reduced using an iris, and as the buffer gas pressure was reduced. These observations can be understood in terms of the fixed position and spatial extent of the fluorescence observation region. When the laser beam is larger than the observation region, the barium atoms are more likely to be pumped into the D states when they are outside the observation region, whereas for a smaller laser beam diameter pumping and fluorescence occur in the region monitored by the photomultiplier tube (see Fig. 2). Similarly, a reduction in buffer gas pressure increases the diffusion rate of Ba atoms

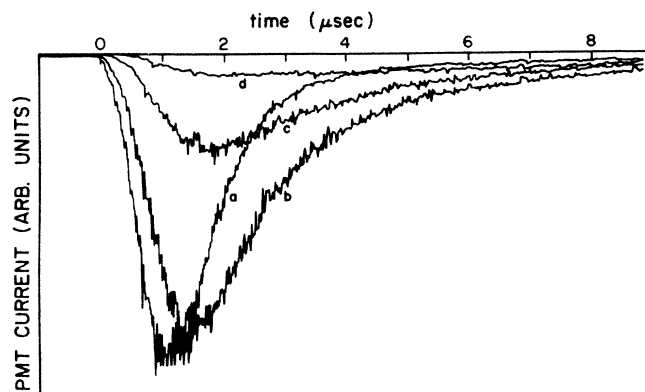


FIG. 3. Examples of Ba fluorescence decay curves, obtained at a buffer gas pressure of 100 torr Ar used to determine the rate of pumping to the metastable states. The curves shown correspond to the following approximate laser intensities (in mW/cm^2): (a) 600, (b) 200, (c) 60, and (d) 20. Note that the vertical scale is inverted.

which penetrate into the observation region prior to being pumped to the D states. At the higher laser intensities used in the experiment, the $^1S_0\text{-}^1P_1$ transition was well saturated, and this explains why in this case, the fluorescence reached its maximum faster than the laser pulse.

To relate the falloff of the fluorescence to the spontaneous decay branch of the 1P_1 to the 1D , 3D levels we use a simplified model in which spatial variations are averaged over the interaction and detection volumes. In this case we may write the following set of approximate rate equations:

$$dN_0/dt = -(R + D)N_0 + (R/3 + A)N_1 + S,$$

$$dN_1/dt = RN_0 - [R/3 + A(1 + b) + D]N_1,$$

$$dN_2/dt = N_1Ab - DN_2,$$

where N_0 , N_1 , and N_2 are the populations of the $6s^2\ ^1S_0$, $6s6p\ ^1P_1$, and the $6s5d\ ^1D$, 3D levels, respectively: R is the excitation rate $^1S_0\text{-}^1P_1$, A is the spontaneous decay rate $^1P\text{-}^1S_0$, Ab is the spontaneous decay rate $^1P\text{-}^1,^3D$, D is the average rate of diffusion of atoms into the laser beam, $S = D(N_0 + N_1 + N_2)$ is a source term representing the rate at which Ba atoms enter the laser beam from the unpumped

TABLE I. Theoretical and experimental results for the $6s6p\ ^1P\text{-}6s5d\ ^1D$, 3D decay rates in BaI.

	1D_2	Spontaneous decay rates ($\times 10^4\ \text{s}^{-1}$) from 1P_1 to		Sum
		3D_2	3D_1	
Theory				
McCavert and Trefftz (Ref. 7)	19	16	0.041	35
Hafner and Schwarz (Ref. 8)	4.6	12	0.12	16.7
Experiment				
Miles and Wiese (Ref. 9)				490
Bernhardt <i>et al.</i> (Ref. 3)				$< 17^a$
Kumar <i>et al.</i> (Ref. 10)				40(4)
This work				210(40)

^aBy considering the decay into the 3D_2 level, which had been neglected in Ref. 3, Niggli and Huber (Ref. 11) estimate a value of $\sim 50 \times 10^4\ \text{s}^{-1}$ from the data in Ref. 3.

volume, and the factor of $\frac{1}{3}$ in the first two equations takes account of the three m states of 1P with complete collisional mixing assumed.

Solving these equations for N_1 , making the approximations $b \ll 1$, $D \ll R$, we obtain for the fluorescent decay constant due to production of metastables

$$K = Ab\left(\frac{4}{3} + A/R\right)^{-1} + D.$$

Further, under our conditions, D was estimated to be of the order of 10^3 s^{-1} and could be neglected. This rate was estimated by using the expression for the lowest spatial mode for the diffusion time for a cylindrical cell⁶ and a diffusion constant for Ba in argon of $0.2 \text{ cm}^2/\text{sec}$. By fitting of exponentials to data such as those shown in Fig. 3, a maximum value of K of $(1.3 \pm 0.1) \times 10^6 \text{ s}^{-1}$ was obtained. An extrapolation of all the data to infinite excitation rate R yielded $K_{\text{max}} = (1.6 \pm 0.3) \times 10^6 \text{ s}^{-1}$. This latter value implies a total decay rate to the metastables of $(2.1 \pm 0.4) \times 10^6 \text{ s}^{-1}$. A comparison is made with previous theoretical and experimental data in Table I.

DISCUSSION

As Table I shows, our result is significantly larger than other recent experimental results and theoretical estimates. The experiments reported involve different *indirect* techniques to obtain the $^1P-^1,3D$ transition rate and are subject to various sources of systematic error. In our experiment, which involves cw pumping of an optically thin Ba vapor in a buffer gas, other mechanisms by which atoms might be transferred to the metastable states or otherwise lost from the system were considered.

Stimulated emission on the $^1P-^1,3D$ transitions has been observed in resonantly pumped Ba vapors at high densities ($N > 10^{15} \text{ cm}^{-3}$) by several workers, e.g., see Ref. 1. However, this cannot be a significant process for vapors which are optically thin ($N < 10^{12} \text{ cm}^{-3}$). As regards collisional

processes, as an alternate explanation for the fluorescence decay, the measured rates would imply surprisingly large cross sections ($\sim 10^{-17} \text{ cm}^2$) for the quenching of 1P to 1,3D by a rare gas. By comparison, measurements of quench cross sections of the $6s5d^1,3D$ metastable states to the ground state by rare gases have yielded values orders of magnitude smaller,^{12,13} consistent with the low residual steady-state fluorescence observed in our data, Fig. 3. Likewise, if chemical gettering by impurity molecules of barium atoms in the 1P excited state were significant, this would imply a very large cross section ($> 10^{-13} \text{ cm}^2$) for the relevant process (impurities in the argon buffer gas stream were believed to be less than 0.01%). Further, it was found that the deliberate addition of approximately 5 torr of hydrogen into the argon stream (by a separate inlet tube so as not to "poison" the barium chip), reduced the observed fall off of the fluorescence by producing a large enhancement of its steady-state value. This is consistent with quenching of the metastables to the *ground state* with a cross section of the order $5 \times 10^{-17} \text{ cm}^2$, in order-of-magnitude agreement with the result in Ref. 13, but not consistent with a much larger cross section for quenching of the $6s6p^1P$ state to the *metastable* states.

In conclusion, we see as the most likely explanation for our observations, the existence of a large spontaneous emission rate to the metastable states. Direct measurement of the decay branch or determination of specific systematics in any of the experiments reported are necessary to resolve the remaining discrepancies. It might also be useful to perform time-resolved experiments with the collision-free environment of an atomic beam.

Note added in proof. Additional theoretical values for the $^1D-^1P$ decay rates have been recently published in Ref. 14.

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