Time-dependence study of radiation trapping by time-delayed two-photon absorption

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The transport of resonance radiation through an optically thick vapor of Sr atoms is studied. A pulsed dye laser tuned to the 461-nm resonance line excites a narrow (~ 250-μm-diameter) column of Sr atoms along the axis of a cylindrical oven containing Sr vapor and Ar buffer gas. After a delay of ≤ 80 ns, a second dye laser excites the atom from the first excited state ($5s5p$) to a higher excited state ($5s7s$). The fluorescence from this latter transition is monitored as the second laser is translated parallel to the first. Since the excited-state–excited-state fluorescence is not trapped, the result is a plot of density of atoms in the $5s5p$ state as a function of position from the originally excited volume. The results are discussed qualitatively.

INTRODUCTION

The transport of energy from an initially excited group of atoms through an optically thick medium of ground-state atoms has been studied since the 1920s.[1] The problem is important in the study of radiative transfer in stellar atmospheres[2] and in laser-produced plasmas.[3] Radiation trapping is also important in a variety of experimental situations in which it is usually an unwanted complication. Consequently, the problem has been extensively studied theoretically (see Refs. 4–8 and references therein).

A number of experimental studies have also been published. Some of these experiments could be classified as steady state. The spectral properties of the scattered light are analyzed when a sample is illuminated by a time-independent white light source[9,10] and a self-reversed profile is observed. Other experiments can be classified as time dependent. In these measurements the decay of the fluorescent light emerging from the sample is observed after the exciting source is turned off. This latter group of experiments can be further subdivided according to whether the sample was in the steady state prior to turning off the excitation source,[1,11–13] or whether the sample was excited by a fast pulsed source.[14–16] In either case an effective lifetime of the resonance radiation is measured.

In all the above experiments it is the resonance light emerging from the cell that is observed. By the very nature of the process this emerging light has been repeatedly scattered and one must attempt to infer from a rather small amount of information the details of a very complicated random walk process. In this paper we present an experimental technique by which we can observe the time-dependent excited-state population as a function of position inside the trapped sample. We can thus probe the dynamics of the trapping process in far more detail than previous studies.

EXPERIMENTAL TECHNIQUE

In Fig. 1 we illustrate the experimental technique. One excimer laser pumped dye laser is tuned to the 460.7-nm resonance line ($5s^2\,{}^1S_0$–$5s5p\,{}^1P_1$) of Sr. This 5-ns laser pulse is directed down the axis of a cell containing Sr vapor and Ar buffer gas. The beam is focused into the center of the cell by a 23-cm focal length lens, L_1, so that near the center of the cell a thin (~ 250-μm diameter) column of excited Sr atoms is created. The laser intensity is not large enough to burn through the entire length of the cell by saturation at line center. However, since the laser linewidth (~ 6 cm^{-1}) is much larger than the Doppler width (~ 0.05 cm^{-1}), the vapor is optically thin in the wings of the laser line; thus a uniform population is excited in the center of the cell by off-resonance absorption from the line wings and by collisionally aided absorption in the line wings.

A second dye laser pumped by the same excimer laser is tuned to the 596-nm transition from the first excited state to the $5s7s\,{}^1S_0$ state. This laser pulse was delayed for up to 85 ns simply by propagation through air. After the delay the second laser beam entered the cell. The two beams coincided at the dichroic mirror, D, which was placed at the front focal point of L_1. Thus by rotating the dichroic mirror with a stepping motor about an axis through the front focal point of L_1, the second laser beam could be translated in the cell parallel to the first beam.

The cell was constructed from a 2 cm inside diameter stainless-steel tubing in the shape of a cross. Each leg of the cross was 20 cm long, the central 10 cm of which was heated. Each leg was fitted with a water-cooled window. A small, spearately heated sidearm in the center of the cell held the Sr. The sidearm temperature was kept about 30 °C lower than that of the main part of the oven.

The density of Sr atoms in the interaction region was estimated in several ways. The density could be found from

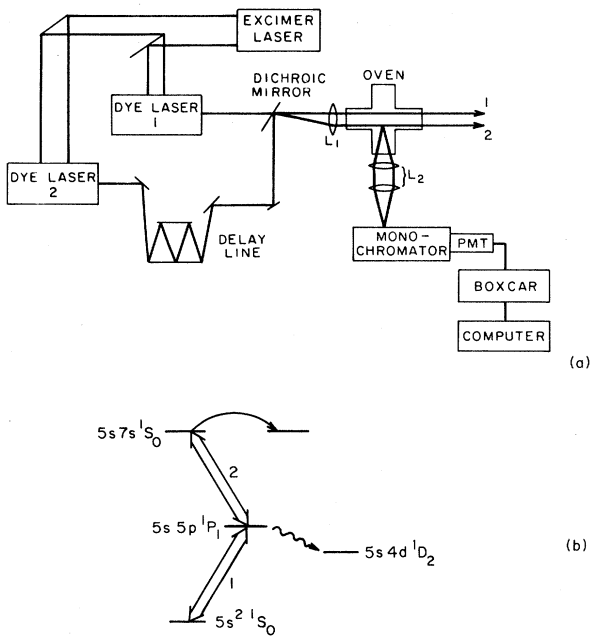

FIG. 1. (a) Block diagram of the apparatus. Dye laser 1 excites a pencil of Sr atoms along the cell axis. Dye laser 2 probes the excited state after a delay time ≤ 84 ns. Fluorescence from the $5s7s$-$6s5p$ transition is detected. (b) Partial energy-level diagram of Sr showing the levels of interest for this experiment.

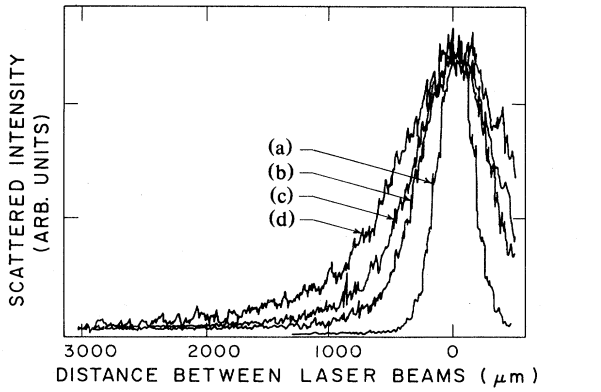

FIG. 2. Typical set of experimental traces showing the $5s7s$-$5s5p$ fluorescence vs position of the second laser beam for delay times of (a) 0, (b) 28 ns, (c) 56 ns, and (d) 84 ns. For all cases the Sr density was 1.2×10^{13} cm^{-3} and the Ar buffer pressure was 35 Torr. The four traces have been normalized to have the same height at the peak.

vapor pressure curves using the sidearm temperature. Also, we made equivalent width measurements of the absorption line.[17] Finally, as we will see below, the experiment itself provides us with an estimate of the absorption coefficient which can be used to obtain the density. When the Ar buffer pressure was below 35 Torr the three density estimates agreed well. At higher buffer pressure the equivalent width measurement yielded lower densities than the other two measurements. We believe the reason for this is that at high pressures the Sr atoms were confined to the central 4–5 cm of the cell thus shortening the effective absorption length through the cell. We therefore have taken as the Sr density near the center of the cell that from vapor pressure data.

Another lens system L_2 collects the light emerging from the center of the cell through one of the sidearms of the oven and focuses it onto the slit of a monochromator. The monochromator is tuned to collect the fluorescence (and Rayleigh scattered light) near 596.0 nm. Because light near 596.0 nm is not trapped the signal at the monochromator output is proportional to the density of excited atoms at the location of the second laser.

The light at the output slit of the monochromator is detected with a photomultiplier and a Princeton Applied Research model PAR-162 boxcar averager. With the time delay of the second laser fixed, its position is scanned and the 596-nm fluorescence as a function of the 596-nm laser position is recorded by computer. Several experimental runs are averaged.

RESULTS

In Fig. 2 we show several typical averaged runs for various time delays. For all these traces the oven temperature was 460 °C and the argon buffer pressure was 35 Torr. The Sr density was estimated to be 1.2×10^{13} atoms/cm^3. The inverse of the line-center absorption coefficient (Beer's length) for these conditions is calculated to be ~ 24 μm. A crude estimate of the behavior to be expected can be obtained by assuming the photons diffuse with a single mean free path given by the Beer's length. The width of the distribution should be $\sqrt{t/\tau}\times 24$ μm, where $\tau \approx 5$ ns is the excited-state lifetime. This qualitative behavior is borne out in the curves of Fig. 2. The width of the zero-delay curve can give us some indication of the spatial resolution of our system. Actually, the system resolution is somewhat narrower than the width of the zero-delay curve because that curve is the convolution of the initially excited region (diameter of first laser beam) with the system resolution. We thus estimate the spatial resolution to be < 250 μm.

The four curves in Fig. 2 have been normalized for illustrative purposes to have the same height at the center. In fact, the signal decreased substantially at the longer time delays. There are two main reasons for this decrease. As the region of excited atoms spreads, of course, the signal will decrease simply to maintain the total number of excited atoms constant (only a negligible fraction of the photons can actually escape the cell in the times of interest here). But the most important reason for the decrease in signal at longer delay times can be seen in Fig. 1(b). The $5s5p$ $^1P_1$ state has an allowed transition to the metastable $5s4d$ $^1D_2$ level. The radiative branching ratio to this level is $\sim 5\%$.[18] Since the lifetime of the $5s5p$ $^1P_1$ level is ~ 5 ns,[18] this means that the total number of excited atoms should decay with a lifetime of ~ 100 ns. To check this we shut off the second laser, tuned the monochromator to observe the resonance radiation, and monitored the time dependence with a Tektronix 5912 transient digitizer. A typical result is shown in Fig. 3. For this case the decay constant is ~ 100 ns, as expected. However, for different temperatures both shorter (~ 50 ns) and longer (~ 200 ns) decay constants were observed. In addition, the signal from the delayed two-photon absorption consistently decayed somewhat faster than would be expected if we only had a simple radiative decay to the $5s4d$ $^1D_2$ state. We have not investigated thoroughly the

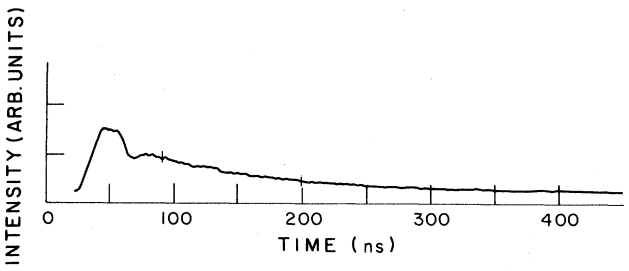

FIG. 3. Decay of the $5s5p$ fluorescence as a function of time. The Sr density was $\times 10^{12}$ and the Ar buffer pressure was 20 Torr. The decay constant of ~ 100 ns was due to collisional and radiative quenching of the $5s5p$ state to the $5s4d$ state.

causes of these observations. Several factors are probably important, including (1) a large uncertainty in the branching ratio, and (2) collisional quenching of the $^1P_1$ state.[19] Future experiments are planned using Mg in which both the loss of signal and the additional complication due to the $^1D_2$ state are absent. Our chief purpose here is to demonstrate our technique and to show the information that can be gained from it.

We have obtained data similar to that in Fig. 2 for Sr densities from $\sim 10^{12}$ to 10^{14} cm^{-3} and for buffer pressures from 1 to 75 Torr. The lowest Sr density which could be used was limited by instabilities in the cell at low temperature. The highest usable density was limited because at 10^{14} cm^{-3} the excited-state density spread very little before decaying to the $5s4d\ ^1D$ state. In Figs. 4 and 5 we show two other sets of curves similar to those given in Fig. 2 but for different values of the Ar buffer gas pressure. If the thermal motion of the atoms is neglected, the excited-state population is expected to spread more quickly at higher buffer pressure because of the increased probability that a photon will be emitted in the Lorentzian line wing where the absorption coefficient is much smaller than at line center. In Fig. 4, in which the buffer pressure is 75 Torr, we clearly

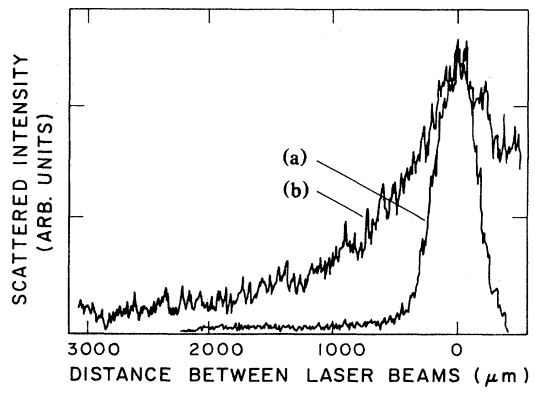

FIG. 4. Fluorescence from the $5s7s$-$5s5p$ transition under the same conditions as in Fig. 2 except that the buffer pressure is 75 Torr. The delays are (a) 0 and (b) 28 ns. The population spreads more quickly than in Fig. 2 because of increased emission in the Lorentzian line wing.

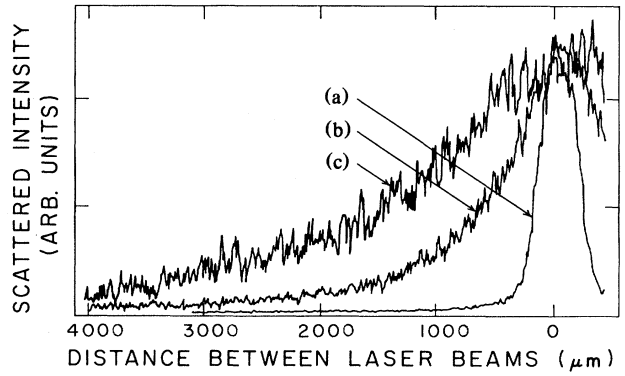

FIG. 5. Fluorescence from the $5s7s$-$5s5p$ transition under the same conditions as in Fig. 2 except that the buffer pressure is 10 Torr. The delays are (a) 0, (b) 28 ns, and (c) 56 ns.

see this effect. At only 28 ns delay, the excited-state population distribution is as broad as the distribution at 84 ns at 35 Torr (Fig. 2). However, in Fig. 5, when the buffer pressure is decreased to 10 Torr, the distribution does not spread more slowly than at 35 Torr; rather it spreads faster. We discuss the interpretation of this result below.

DISCUSSION

Because of the loss of population in the excited state in a relatively short time ($\lesssim 100$ ns) some interesting features of the general trapping problem do not occur in the Sr system. These include effects due to the boundary conditions and to incomplete redistribution. We have, however, been able to observe the transition in the trapping problem mentioned above for which we have no quantitative or even specific qualitative explanation. We shall content ourselves with a discussion of relevant parameters and some suggestions about particular mechanisms that may be involved in the transition.

Once excited, a strontium atom will move in a straight line until it either emits a photon, suffers a velocity changing collision with an Ar atom, or exchanges excitation through a resonance collision with a ground-state Sr atom. In any case the excitation then moves off with a new velocity. Consequently, the excitation diffuses not only due to the random walk of the photon but also due to the thermal motion of the excited atoms: could this play a role?

To estimate the relative importance of these processes we first estimate the collision rate for momentum changing collisions and for excitation transfer. We assume a cross section of 10 Å^2 for momentum changing collisions. At a buffer pressure of 10 Torr we obtain a collision rate of $\sim 1.5\times 10^7$ s^{-1}. The cross section for resonant energy transfer has been calculated to be $\sim 9\times 10^{-12}$ cm^2.[20] For a Sr density of 1.2×10^{13} cm^{-3} the collision rate is 5.5×10^6 s^{-1}. Thus, during even the longest times of interest in this experiment an excited atom will suffer few of these types of collisions under these conditions. To discuss the effect of atomic motion we can therefore assume that the atom simply moves in a straight line trajectory with velocity

$\sim 4.2\times10^4$ cm s^{-1}. In one lifetime (5 ns) a Sr atom would move about 2 μm. This is far less than the distance the excitation should travel in this time due only to photon hopping, the Beer's length for the problem being 25 μm. At higher Sr and/or Ar pressures the collision mechanisms discussed will impede the thermal motion but we do not believe this to play a major role in the overall trapping problem (Fig. 2).

In the region where the transition occurs the radiative and collisional rates are approximately equal. This means that partial redistribution—both coherent and incoherent scattering—must be considered. It would seem likely then that the onset of coherent scattering is playing a major role. This onset, however, can manifest itself in several ways, firstly, in an angle averaged sense, purely in terms of preservation of frequencies. It seems more likely, however, that the preservation of photon propagation direction, and perhaps even polarization, will also be involved. Preservation of the propagation and polarization in the low-density limit may well change the evolution of the excitation from a diffusive to streaming behavior.

We should stress that the above arguments are for the purpose of establishing the processes which must be considered in the analysis of these data. We know of no theory that is able to quantitatively fit these results. Payne and Cook[6] have presented explicit solutions to the Holstein equation for cylindrical geometry in the short-time limit but they have not included atomic motion or partial redistribution. Hummer and Kunasz[8] have shown how to include atomic motion but have only presented results for a slab geometry. Atomic motion has also been discussed by Cippola and Morse.[21] Again all these studies assumed complete redistribution.

Clearly, it would be desirable to have a theory with which to compare these results, and progress is being made in this direction.[22] However, the experimental data are also quite limited largely as a result of quenching of the excited state to the $5s4d$ level. Futher experiments are planned using Mg rather than Sr in which quenching should not be a problem. Preliminary experiments have indicated that measurements can be made at delays of at least 1 μs in Mg. Such long delays are obtained using a fiber optic delay line.[23] For future experiments also a new cell design will be used in which the boundary condition can be well controlled and the vapor density can be well known and very uniform. We expect to be able to study many aspects of the radiation trapping problem with this technique. We shall, in particular, attempt a detailed elucidation of the mechanism of the transition we have observed through a systematic variation of the relevant parameters.

CONCLUSIONS

We have demonstrated a technique for studying time-dependent resonance radiation trapping in far more detail than previously possible. A number of improvements of this system are planned so we can quantitatively compare our results to trapping theories. As we have already mentioned, the use of Mg rather than Sr will eliminate complications due to the ^{1}D state and will allow longer delays to be observed. For quantitative comparison with theory a well-controlled geometry is necessary. Thus a uniformly heated cylindrical cell with no sidearms will be used.

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