EXPERIMENTAL ASPECTS OF STUDYING THE POLARIZATION OF COLLISIONALLY
REDISTRIBUTED LIGHT IN STRONTIUM-RARE GAS SYSTEMS

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INTRODUCTION

The past several years have seen a significant interest in the problem of
collisional redistribution of radiation. Early theoretical work in line
broadening was concerned with explaining absorption (or emission) data in
terms of intermolecular potential curves. This led, for example, to using
quasi-static line broadening theory and experimental data to obtain dif-
ference potentials [1]. Absorption (or emission) experiments of this type
are rather straightforward to perform and have been done for a large num-
ber of systems [2]. In cases of temperature dependent absorption (or
emission) data one can obtain the actual intermolecular potential curves
for the individual states involved in a transition [3], rather than just
the difference potential. Temperature dependent absorption experiments,
though, are more difficult than simple absorption experiments and less
common in the literature. Recent theoretical work has been more complete
and allows one to consider far wing scattering experiments [4]. The hope
of obtaining detailed information about collision dynamics via the redis-
tribution process has led to the recent scattering experiments.

One type of scattering experiment is to excite, during a collision, an
atomic system with linearly polarized light tuned near (1-1000 cm\(^{-1}\)), but
not on, resonance and observe the polarization of the fluorescence. Ex-
periments of this type are conceptually easy but offer some experimental
difficulties not found in absorption experiments. We have performed such
an experiment and wish to discuss in some detail the technical problems
associated with it. We shall give an example of the data one can obtain
from the experiment and discuss future directions for work. Our complete results will be discussed elsewhere [5].

DESCRIPTION OF EXPERIMENT

Linearly polarized light tuned near the resonance line of Sr (4607 Å, J=0 to J=1) being perturbed by Ar was used to excite the Sr. The fluorescence was observed 90° to the incident direction of propagation. Measuring the components of the fluorescence parallel and perpendicular to the incident polarization direction allows one to calculate the polarization of the fluorescence. The polarization is defined as the difference of the components polarized parallel and perpendicular to the incident polarization direction over the sum of the two: \[ \text{Pol} = \frac{(I_\parallel - I_\perp)}{(I_\parallel + I_\perp)}. \]

The set-up for the experiment is very similar to that used by Thomann et al. [6], see Fig. 1. The uv lines of a Coherent CR-15 Ar⁺ laser are used to pump a Coherent 590 dye laser using Stilbene-3 laser dye. Typical output of the dye laser is 50 mW and is linearly polarized. It is also possible to use the blue Ar⁺ laser lines directly. This allows us (due to higher intensity) to go much further into the wings than the dye laser alone would allow. After monitoring the laser power and wavelength this light is sent into a heated cell containing Sr vapor and Ar buffer gas. Rather than send in one polarization and observe two perpendicular polarizations, one can send in two perpendicular polarizations and observe just one. This is done because the laser polarization direction is easily controlled with a Pockels cell. Specifically, light polarized in the z and x directions is used for excitation while light polarized in the z direction is observed. The fluorescence is spectrally resolved from the laser wavelength with a 1.0-m monochromator. Low light signals require use of a cooled photomultiplier and a computer controlled photon counting system.

The cell is a stainless steel cross which is heated near the center and cooled at the ends of the arms. The cell is not completely in thermal equilibrium. This means there can be slow Sr density fluctuations.
However, the computer takes data fast enough that the density fluctuations are not a problem. Typical cell temperatures are 450 C-600 C. An optically thin path for the fluorescence is needed since the polarization of the fluorescence is affected (reduced) by multiple scattering. To accomplish this a heated sapphire window is placed in the interaction region, see Fig. 2, and the Sr vapor pressure is set sufficiently low (see below). Passing the laser very close to the sapphire window reduces the vapor path length of the fluorescence to approximately 150 microns. The sapphire window and the cell are kept at temperatures higher than the finger (see Fig. 2) so the Sr does not condense on the sapphire window. A lens is placed close to the interaction region to increase the signal collected.
Fig. 2. Cell interior. This is a cross-sectional view of the center of the cell. The laser propagates perpendicular to the plane of the figure.

The sapphire window assembly is mounted on the cell and extends into the interaction region. The assembly is made of Kovar and the sapphire window is press sealed with copper gaskets (on both sides of the window) to the Kovar. The sapphire-Kovar seal is very similar to that of Zajonc [7]. This is not a high vacuum seal but it keeps any Sr vapor out of the region behind the sapphire window. Sapphire is birefringent so the window must be carefully oriented. The window contains two perpendicular axes which do not affect linearly polarized light parallel to these axes. The window must be oriented such that one of the neutral axes is parallel to the direction of polarization being observed.

As stated above, the purpose of the sapphire window is to stop multiple scattering of the fluorescence. To verify that multiple scattering is not a problem the polarization as a function of finger temperature (the finger temperature determines the Sr density) is observed. Figure 3 shows results for Sr-Ne. Sr-Ar results are very similar. The polarization rises and levels off as a function of decreasing Sr density. Detuning dependent polarization data was taken on the flat part of the curve. This is an
Fig. 3. Polarization vs. finger temperature. The finger temperature determines the Sr density in the interaction region.

An important test for an experiment of this kind because it assures us of no multiple scattering and helps rule out any effects due to resonance broadening.

RESULTS

Results for Sr-Ar are shown in Fig. 4. The Sr density was approximately $10^{13}$ cm$^{-3}$ and the Ar pressure was 3 torr. The impact region for Sr-Ar extends to about 3.6 cm$^{-1}$ (inverse of collision time). The red wing curve as a function of increasing detuning shows an initial drop in polarization past the impact region. A valley at 33% is then observed from 15 cm$^{-1}$ to 40 cm$^{-1}$. Further out in the wing the polarization rises to 45% at 500 cm$^{-1}$. The blue wing shows a drop in polarization past the impact region and a leveling off at large detuning. The interpretation of these results has been considered by Cooper [8] and Lewis et al. [9].
Fig. 4. Polarization curves for Sr–Ar. Data were taken at an Ar pressure of 3 Torr.

FUTURE EXPERIMENTS

We set out to do this experiment because the formal theory [4] led us to believe a light scattering experiment could be used to study the details of mixing of atomic states during a collision. To date, semiclassical models [8,9] and fully quantum mechanical calculations of ab initio potentials [10] have been used to explain qualitative results of depolarization data. These studies confirm our suspicion that depolarization data are critically dependent on intermolecular potential curves and nonadiabatic mixing during a collision. Thus, we believe data of this type provide the molecular theorist with new and strong constraints on molecular collision calculations.

Depolarization data contain information on the propagation from the point of absorption to the asymptotic states. More information on the details
of this propagation would be obtained though, if we could observe the
light emitted during the same collision; for this would involve propaga-
tion between two points on the potential curve. This process is, however,
rather unlikely: only \( \sim 10^{-4} \) of the atoms re-emit in the same collision.
We can, however, look for induced absorption to a higher-lying state which
occurs during the same collision. This may be preferable since we can
induce the second absorption with, say, a second, tunable laser. Thus,
by varying the frequency of both lasers we obtain information on point-to-
point propagation during a collision. Just how much more physics one can
extract in this situation is an open question. The general theory sug-
gests that this should be an even more powerful tool than the collision
induced fluorescence experiments. An analysis of this type of process
has been made by Berman and Yeh [11] and the effects of degeneracy can be
handled using the techniques of Burnett and Cooper [4]. Experimental and
theoretical work on this problem is now under way.

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