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OBSERVATION OF LUMINESCENT SPECTRA
IN LOW-ENERGY ION-NEUTRAL COLLISIONS

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J. J. Leventhal

University of Missouri - St. Louis
St. Louis, Missouri 63121

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ABSTRACT

This report describes work performed during the past year. Experiments on energy pooling in excited atom/excited atom collisions and laser enhanced charge transfer were performed and reported in the scientific literature. Work continued on these aspects of the program as well as on experimental studies of trapping of resonance radiation.

INTRODUCTION

This report describes work done in the past year in the Atomic and Molecular Physics Laboratory at UMSL. The primary objective is to perform experiments designed to obtain data on energy transfer and energy partitioning in inelastic atomic and molecular collisions. A variety of techniques are employed including emission spectroscopy and atomic beam methods.

During the past year progress was made in three aspects of this program:

1. Laser-assisted charge transfer
2. Energy pooling in excited atom/excited atom collisions
3. Trapped lifetime experiments

These topics will be discussed separately.

II. Laser Assisted Charge Transfer

These experiments are designed to investigate reaction selectivity based on reactant internal energy. This kind of selectivity is of interest because such processes have been proposed as possible methods for isotope separation. In addition reactions involving excited species have been studied as a potential means of producing population inversions. Our intention has been to study the fundamental nature of such processes by examining simple collision systems. We chose the proton sodium system to begin our work. Although a number of unanticipated experimental problems that hampered progress on these experiments were encountered, studies of H^+-Na were completed during the past year and the results reported in The Physical Review Letters¹.

Briefly, we wish to compare the yield of Lyman α ($L\alpha$) radiation from the reaction

with that from the reaction

Reaction (1) is endothermic by 1.74 eV while reaction (2) is exothermic by 0.34 eV. Since the potential energy curves for $H(n=2)-Na^+$ lie closer to the $H^+-Na(3p)$ curve than the $H^+-Na(3s)$ curve it is expected that reaction (2) will have a larger cross section than reaction (1).

The apparatus for these experiments is shown schematically in Figure 1. A mass selected proton beam of variable kinetic energy (1-2000 eV) is directed into a cell containing sodium vapor at $\sim 10^{13}$ atoms/cm³. For these experiments the cell is illuminated with light from a cw tunable dye laser which can be mechanically chopped. On one side of the cell there is a slot leading to an optical system (monochromator and photomultiplier) for detecting and dispersing collision-produced radiation in the wavelength range 2000-8500. This optical system can be used, as it has been in the past, to obtain collision-produced emission spectra for the purpose of diagnosing inelastic ion-atom or ion-molecule collision processes. It can also be used in experiments involving laser excitation when appropriate; in this case a "notch" absorption filter is used to eliminate most of the intense laser-induced resonance fluorescence radiation which would otherwise damage the photomultiplier tube.

Figure 1. Schematic diagram of the apparatus used in laser assisted charge transfer experiments.

There is a 3 mm. diameter hole on the side of the cell opposite the optical system. This hole may be used to extract ions from the interaction region for mass analysis or, as in the present case, to pass vuv radiation.

The vuv radiation is detected with a solar blind photomultiplier as shown. To select $L\alpha$, a 1216Å narrowband filter is interposed between the cell and the photomultiplier. A dual counter, synchronized with the mechanical light chopper, is used to compare the "laser on" to "laser off" conditions, and thus assess the relative roles of reactions 1 and 2.

Our results show that the expected enhancement occurs, but only for kinetic energies below about 30 eV. We also demonstrated that neither H_2^+ -Na nor H_3^+ -Na collisions produced a measureable enhancement of $L\alpha$ or any radiation at or near the $L\alpha$ wavelength (such as radiation from excited H_2 molecules). Further details are given in Appendix I which is a reprint of the paper in The Physical Review Letters.

III. Energy Pooling

These experiments are designed to investigate the fundamental nature of energy transfer processes involving two atoms or molecules, each of which is in an excited state. Our studies so far have concentrated on reactions of the type

and

For both Reactions (3) and (4) the indicated products are formed by "pooling" the internal energy of two 3p sodium atoms. In fact, in Reaction (3) if the $n\ell$ sodium state lies higher than twice the 3p energy (2.1 eV) then kinetic energy is also required to drive the reaction. Thus "energy pooling" reaction in this context could refer to a process that requires a combination of internal energy from both reactants and possibly conversion of reactant kinetic energy into internal energy to access the products.

After our initial report of our observation of such processes² we began a systematic study of Na(3p)-Na(3p) collisions. The apparatus used in these experiments is shown schematically in Figure 2.

Figure 2. Schematic diagram of the apparatus used in energy pooling experiments.

A Spectra-Physics Model 580 single frequency dye laser pumped by a 5 watt Ar^+ laser, and tuned to either of the sodium D_1 or D_2 lines is used to excite sodium atoms to the $3p$ state. A small fraction of the laser beam is directed into a 2GHz FSR, 13 MHz bandwidth confocal interferometer, Spectra-Physics Model 470-3, and displayed on an oscilloscope to monitor frequency stability. The main laser beam is focused into the collision cell and the laser power density varied using a set of calibrated neutral density filters.

In addition to the apertures for the laser beam, the collision cell has a 2 cm long X 2 mm wide slot parallel to the laser beam. Photons emanating from the collision cell that pass through the slot may be dispersed with a 0.25 m. scanning monochromator. A notch filter with transmission 10^{-7} at 589 nm is used to attenuate the 589 nm fluorescence from the sodium vapor. However the transmitted signal is usually high enough to detect and provides a convenient monitor at the $\text{Na}(3p)$ population. After passing through the monochromator, photons are detected with a cooled photomultiplier tube used in the counting mode. Ions formed in the laser-excited vapor are collected on an electrode inserted into the collision cell, and the ion current measured with an electrometer. A small transverse electric field ensures complete ion collection. Although both Na^+ and Na_2^+ ions are known to be formed in the laser-excited vapor, the origin of the Na^+ has been determined to be photodissociation of Na_2^+ so that the total ion signal is a measure of incipient Na_2^+ .

Sodium vapor is introduced into the collision cell from an oven located beneath the cell. This configuration is necessary to minimize blackbody radiation in the optical system. For this reason the reading on a thermocouple mounted in the oven cannot be used to infer the sodium atom

density in the cell. Instead, the atom density, n , is presently determined by directing a mass selected N_2^+ ion beam into the cell, and observing the 589 nm radiation resulting from collision induced excitation in N_2^+ -Na collisions. This signal, together with the previously measured absolute cross section for this N_2^+ -Na³ process, leads to a determination of n as a function of the oven thermocouple reading.

Experiments performed within the past year were directed toward verifying the processes responsible for the observed products, and determining rate constants for these processes. Temperature dependences were also investigated. The results of this work was reported in The Physical Review⁴; a reprint of the paper is included in this report as Appendix II.

IV. Trapped Lifetime Experiments

This work was begun with the anticipation that it would lead to an alternative to those methods currently employed for determining atom densities in metal vapor. Because our collision cell is cooler than the oven from which the Na atoms emanate, and because the cell has openings for ion extraction, the temperature of the oven cannot be correlated with the atom density, n . As noted in the previous section of this report we use ion impact on sodium atoms in conjunction with a previously measured cross section for the inelastic process

to determine n in our energy pooling experiments. The "previously measured" cross section was determined using a surface ionization detector. For experiments involving other alkali atoms processes analogous to Reaction (5) are employed.

Our new method is based on the well-known effect of imprisonment of resonance radiation. It has long been known that the measured lifetime, τ^* , of an atomic (or molecular) state can be longer than the natural lifetime, τ , of the state if the time decay of a resonance line is used for the lifetime measurement. Since the final state of a resonance line (in emission) is the atomic ground state, nearby atoms in the ground state may reabsorb emitted resonance radiation. Thus, observation of resonance radiation emitted by excited atoms may be delayed due to this trapping effect. The apparent lifetime then is expected to be a function of the atom density and the