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A Spectroscopic Study of Hydrogen Atom and Molecule Collisions

Final Report

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1 Introduction

In this project the fundamental processes which occur in low energy collisions of excited states of atomic hydrogen with other atoms and ions are being studied with optical, vacuum ultraviolet, and laser spectroscopy. This final report covers work through June 2002. We begin here with a brief description of the status of the work at the beginning of the last project period in 1997, then discuss the goals for this last period and our results.

The purpose of our work is to understand low energy atom-atom collisions during which light is emitted or absorbed. Because of their fundamental character, such collisions of atomic hydrogen could play a central role if experimental data could be compared with *a priori* theory. Some interactions involving atomic hydrogen can be calculated very accurately, namely those of H_2 , H_2^+ , H_3 , and H_3^+ , and simpler diatomic radicals. The primary difficulty from the experimental side has been the development of techniques to observe neutral atomic hydrogen interactions at densities high enough for spectral line broadening effects to be observable, to characterize quantitatively conditions under which the spectra are created, and to distinguish other sources of continua from those arising through atomic collisions.

We use a laser-produced plasma in a static gas target as a source of atomic H and H^+ , to explore the Lyman α wing above 1215 Å. A model of the plasma production process has been developed, and we have a basic understanding of the shock wave physics which leads to the dissociation of H_2 and production of atomic H. A diagnostic tool, shadowgraph imagery, has been applied to show the evolution of the post-plasma bubble. Images of the laser focus and the plasma have revealed channel formation, and evidence of filamentation at higher pressures. The spectrum exhibits features in the far wing of Lyman α that are identified as those expected on the basis of line shape theory for atom-atom and atom-ion collisions. In collaboration with N. Allard of the Institut d'Astrophysique, we also improved the framework of the theoretical model, and developed methods for computing the contributions from all possible molecular states of H_2 and H_2^+ to the spectrum of free atoms in collision. Experimental work also identified where and when H_3^+ appeared in the laser-produced plasmas, since it is an important species in the chemistry of the plasma when other atoms are added to create radicals such as CH and OH. We have detected OH both in emission and in fluorescence as a post-plasma product when H_2O was seeded into the initial atmosphere of otherwise pure H_2 .

The goals of the project during the final period were designed to build on this foundation. With the contributions from several graduate students, we worked to refine the plasma production model and include in it the role of the molecular dissociation in more detail, in order to follow the dissipation of the shock through the phase which leaves a static bubble in the surrounding gas. We identified OH emission, and characterized the state of the radical long after the visible plasma had disappeared. We also detected a continuum from OH that may be a signature of radiative dissociation from a highly excited state. We made low resolution measurements of the known H_2 continua from a positive column discharge, for comparison to continua seen from the laser-plasma. We made improvements to our 5-meter vacuum spectrometer to improve vibration isolation and vacuum quality; this instrument is used in this project to measure line spectra, distinguish continua from unresolved lines, and measure line "core" regions accurately. Measurements of the Lyman α wing from a laser plasma in a static cell were made for comparison with the line shape theory. These measurements have now been extended to include initial pressures of up to 100 atmospheres. Significant improvements to the theory were made as well, which allowed systematic inclusion of the full complexity of states that result from interactions of excited atomic H with other H and H^+ atoms and ions. The molecular structure, branching ratios, statistical weights, variation of radiative transition moment with interatomic separation, and ensemble statistics in the source are now included. Evaluations of the theory were made for Lyman α and β , γ and Balmer α in support of the experimental work.

A brief review of the progress which has been made on the Lyman α experiments and the theory of Lyman α and Lyman β is given in the following sections.

2 Lyman α experiments

When a 1064 nm 6 ns Nd:YAG laser pulse with an energy of the order of 300 mJ is focused into a cell containing H_2 , additional self-focusing of the beam causes most of its energy to be delivered suddenly to a cylinder about 0.5 cm long and 50 μm in diameter surrounding the focal point. As a consequence, a shock wave propagates outward and leaves behind a cooling mixture of neutral H, H_2 , ions and electrons to provide a source for studying radiative collisions. Simple models of the shock excitation, dissociation,

and ionization are consistent with space and time resolved diagnostic spectroscopy of this source. Of particular interest to us is the extraction of the spectral line profile in the vacuum ultraviolet Lyman- α region. Time resolved emission spectroscopy combined with models of the post shock gas allows us to identify features in the Lyman- α profile that are the result of radiative collisions of an excited H atom and an unexcited atom or ion perturber[1]. We describe the experiments here, and compare the observed spectra with new unified theory Lyman α calculations [2].

The plasmas were made inside a stainless steel six-way cross. Fluorescence-free fused silica windows along one axis permitted a laser beam to be focused at the center of the cross and then exit to a beam dump. A lens was located so that its focal point for the laser light was centered in the cross. The target region was pumped by a liquid-nitrogen trapped diffusion pump, and vacuum integrity and the presence of residual gases were monitored with a quadrupole mass spectrometer. Once the clean out-gassed cell was filled with 99.995% purity H_2 , it was used for spectroscopy in a static mode.

The spectrum of the plasma was observed at 90° to the plasma-forming laser beam. In the vacuum ultraviolet the emitted light passed along one axis of the cross, through a MgF_2 window, through an entrance slit, and into an a monochromator with an iridium-coated holographic concave grating. The dispersed spectrum was imaged by the grating onto an exit slit and then to a solarblind photomultiplier. The MgF_2 windows of the photomultiplier and the monochromator absorbed light with $\lambda < 1150 \text{ \AA}$, and the photocathode of the detector was not sensitive to $\lambda > 1800 \text{ \AA}$. Separate calibrations of the detector quantum efficiency and grating reflectivity were combined to correct for a system efficiency, and this was checked by comparing the spectrum of H_2 observed from a positive column discharge with a theoretical spectrum simulation for the molecule.

In the plasma model calculations we assumed an initial pressure of 800 Torr of H_2 at 300 K into which the laser deposited 600 mJ in a 0.5 cm long channel to start the expansion. A cylindrical bubble of hot ionized gas formed behind the shock with a radius which expanded rapidly after the initial laser pulse. Once the expansion was underway and the excitation pulse was off the density was very low close to the axis where the temperature was high. Farther from the axis and just behind the shock front the gas was compressed and the density was greater than ambient. With time the outward flow excavated a cavity in the surrounding gas cell. Neutral atoms in the shock heated volume were on a ridge in the r-t plane which was defined on its outer edge by the

Figure 1: Density of atomic H versus time and distance. The white line marks the boundary of the shock wave.

dissociation of H_2 , and on its inner edge by the ionization of the atoms. The atomic density in this ridge exceeded the ambient molecular density immediately after the laser pulse because of the compression and also because of the dissociation of diatomic H_2 into two atoms. The distribution of H-atoms is shown in the three dimensional representation of Fig. 1. The emission responsible for Lyman α arose in this outwardly moving shell. Inside the shell there was a similar ridge in the electron density, where the outer edge was defined by the ionization of the atoms, and the inner edge by the density gradient of post-shock gas.

Figure 2 compares a line shape calculation for the 1600 Å region observed in the time period when ion densities are low and charged particle collisions produce a negligible effect on the profile. The theoretical profile matches the observed shape of the 1600 Å satellite well. This is an important point because the line shape theory predicts that this satellite is enhanced by an increase in the radiative dipole moment in the region of internuclear separation that contributes to this satellite.

The laboratory observations of a laser-produced plasma confirm that satellites appear on Lyman α due to collisions with neutral atoms and pro-

Figure 2: Comparison of the experimental 1600 Å region with theoretical models with and without variable $D(R)$. The line wing is optically thin in this region. F_ν is normalized to unity for an integral of the line profile over ν in cm^{-1} , and scaled for a neutral H density of $10^{19} \text{ atoms/cm}^3$. The observed shape of the satellite is in good agreement with the variable $D(R)$ theory; the constant D theory underestimates the strength of the satellite. Both theories predict oscillatory structure between the satellite and the line.

tons. This experimental confirmation of the theory supports the identification of these features in the Lyman α spectra of white dwarf and λ Bootis stars. Blastwave models of the plasma are consistent with the observed strength of ion and neutral satellite features in the Lyman α wing. For stellar atmospheres the line shape models have proven useful in diagnostic measurements of ion and neutral densities, and thereby temperature. It now appears that the same is true for laboratory plasmas, and that a comparison of the Lyman α wing with line shape models is a tool for determining neutral and proton densities in a hydrogenic plasma.

These experiments also confirm that the variation of the radiative dipole moment is an important factor in determining the far wing emission of Lyman α . Most previous work on far wing broadening has made the simplifying assumption that $D(R)$ is a constant, but based on this work it appears that assumption may not be valid. We note that when $D(R)$ differs significantly from its asymptotic value at an R close to the region forming a satellite, the strength of the wing may be enhanced (or diminished) considerably. Certainly when satellites are used as density diagnostics for plasmas, this is a factor which needs to be considered [1, 2]

3 Lyman series theory

In Allard *et al.* 1999 [2] we derived a classical path expression for a pressure-broadened atomic spectral line shape that allows for a radiative electric dipole transition moment which depends on the position of the perturbers. This factor is not included in the more usual approximations for neutral atom line shape calculations. We used this theory to study the influence of the variation of the dipole moment on the satellites present in the far wing profiles of the Lyman series lines of atomic hydrogen seen in stars and in laboratory plasmas [3].

Satellite features in hydrogen lines are not limited to Lyman α , which is the only Lyman-series line accessible to the Hubble Space Telescope, and the only one for which we as yet have laboratory data. Observations with HUT (Hopkins Ultraviolet Telescope) and with ORFEUS (Orbiting Retrievable Far and Extreme Ultraviolet Spectrograph) of Lyman β of DA white dwarfs with T_{eff} close to 20000 K have revealed a line shape very different from the expected simple Stark broadening, with line satellites near 1078 and 1060 Å [4, 5]. The satellites in the red wing of Lyman β are in the 905 to

1187 Å spectral region covered by the Far Ultraviolet Spectroscopic Explorer (FUSE) launched in June 1999. Furthermore, Lyman β profiles are also the subject of an ongoing study in this laboratory of the far ultraviolet spectrum of dense hydrogen plasmas. The strengths of these satellite features and indeed the entire shape of wings in the Lyman series are very sensitive to the degree of ionization in the stellar atmosphere and laboratory plasmas, because that determines the relative importance of broadening by ion and neutral collisions.

The recent *ab initio* calculations by Drira 1999 [6] of electronic transition moments for excited states of the H_2 molecule and new accurate molecular potentials of Schmelcher [7, 8] now allow us to compute Lyman β profiles simultaneously perturbed by neutral atomic hydrogen and by protons. In a recent paper we examined a collision-induced satellite correlated to the $B''\bar{B} \ ^1\Sigma_u^+ - X \ ^1\Sigma_g^+$ asymptotically forbidden transition of H_2 [3]. We showed that the shape of the wing in the region between Lyman β and Lyman α is particularly sensitive to the relative abundance of the neutral and ion perturbers responsible for the broadening of the lines.

The sum of the profiles of Lyman α and Lyman β are shown in Fig. 3. When the neutral density is increased by a factor of 5 over the ion density, a collision induced satellite appears in the far wing. This satellite is therefore sensitive to the degree of ionization and may be used as a temperature diagnostic in a plasma where Saha ionization-equilibrium holds. In the case of Lyman α and the $H-H^+$ Lyman β satellites, the potential shape plays a dominant role in the large difference in the broadening of the quasi-molecular features [9]. Here, the width of the collision-induced satellite is determined, for the most part, by the limited region over which its transition dipole moment is significant.

In a collision-induced spectrum such as this one, the dipole moment may be more important than the potential in determining the shape of the satellite. The observation of such a satellite therefore would be a test of the accuracy of the dipole moment calculation and the line shape theory that depends on it. The complexities of the profile shown in Fig. 3 illustrate how important it is to use a complete line shape theory that includes time dependence as a fundamental element. These "finite duration of collision" effects are very significant in the far wing of spectral lines.

Figure 3: Total profile of Lyman α and Lyman β perturbed by neutral hydrogen and protons. Three different neutral densities (1×10^{16} , 5×10^{16} , $1 \times 10^{17} \text{ cm}^{-3}$) are compared for a fixed ion density ($1 \times 10^{16} \text{ cm}^{-3}$).

4 The Balmer Series at High Density

In recent experiments we extended the measurements to higher atomic densities, in order to reveal the effects of several perturbers acting simultaneously on one radiating atom. These many-body perturbations alter the far line wing, adding multiple satellites and producing a strong continuum from the vacuum ultraviolet to the ultraviolet and visible. One way to make these effects more detectable is to use a static high pressure cell, and measurements of laser plasmas at initial molecular gas densities over 100 atmospheres were made to see whether the far wing resembles the predictions of a simplified theory in which the effects of individual perturbers add linearly. We found that the additive approximation which is used in the line shape model provides a reasonable estimate of the strength of this wing, but will require improvement to reproduce the structure representative, for example, of free-free transitions of excited H_3 or of an excited H-atom interacting simultaneously with two free protons and two or more free electrons [10]. These measurements also allowed a determination of the shift of Balmer α at electron and ion densities up to 10^{20} cm^{-3} , an order of magnitude larger than had been achieved in stabilized arc experiments, which should help to resolve questions about the size and origin of the high density shifts in the Balmer series [11].

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