Electron collisions with constituents of planetary atmospheres

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Abstract. Collisions between electrons and neutral species play an important role in the energy balance of upper atmospheres throughout the Solar System. These processes, and the subsequent photon emissions produced, provide one of the primary means for probing, diagnosing and understanding the dynamics of these environments. Modelling of these plasmas and interpretation of optical observations requires accurate knowledge of atomic/molecular parameters such as oscillator strengths and predissociation yields along with cross sections for electron impact excitation (and emission) with the constituent species of these atmospheres. In this paper, our recent work involving electron collisions with two of the most important atmospheric species in our Solar System, namely N\textsubscript{2} and H\textsubscript{2}, will be reviewed. Preliminary differential cross sections for excitation of the B \textsuperscript{1}\Sigma\textsuperscript{u}\textsuperscript{+} and C \textsuperscript{1}\Pi\textsuperscript{u} states out of the X \textsuperscript{1}\Sigma\textsuperscript{g}\textsuperscript{+} (v\textsuperscript{″}=1) ground state level of H\textsubscript{2} are presented.

1. Introduction

In planetary atmospheres, a principal mechanism for energy exchange involves a combination of collision processes between electrons and photons (excitation, ionization, attachment, etc.) with neutral and ionized species. The auroral phenomenon in the Earth’s atmosphere \cite{1}, the electroglow in the ionosphere of the giant planets \cite{2, 3}, ultraviolet (UV) emission from the Io plasma torus \cite{4}, and UV emission from Titan \cite{5} and Triton \cite{6} clearly demonstrate the prevalence of electron impact induced excitation and subsequent fluorescence from atmospheric species in the Solar System. Fundamental properties of species found in planetary plasmas (i.e., collision cross sections, oscillator strengths, predissociation yields, etc.), determined by laboratory and theoretical investigations, are essential to the understanding of observational data through models of these systems. The importance of studying these processes is in part emphasized by the extensive investment to date made in obtaining far ultraviolet (FUV) and extreme ultraviolet (EUV) emission data from various atmospheres throughout the Solar System via spacecraft based UV spectrographs such as the Voyager Ultraviolet Spectrometer (UVS), the Galileo Extreme Ultraviolet Spectrometer (EUVS), the International Ultraviolet Explorer (IUE), the Hubble Space Telescope (HST) Goddard High Resolution Spectrograph (GHRS), the HST Faint Object Spectrograph (FOS), the Hopkins Ultraviolet Telescope (HUT), the Far Ultraviolet Spectroscopic Explorer (FUSE), the Cassini Ultraviolet Imaging Spectrometer (UVIS), etc. However, there exist a number of deficiencies in the

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current atomic/molecular data base, which hinders the accurate interpretation of these difficult to obtain and expensive observational data. In response, our group has developed a program to target these deficiencies. In this paper we discuss recent work related to H₂ and N₂.

2. Vibrationally excited hydrogen (H₂)

Electron impact excitation, dissociation, ionization, and electron attachment play important roles in the production of photon emissions in the UV and in the transport of energy (heating and cooling) in the Jovian and Saturnian systems. In both of these environments, hydrogen molecules in vibrationally excited levels of the ground state, i.e., $X^1Σ^+_g (v'' > 0)$, are in abundance and play an important role in the atmospheric dynamics [7-9]. In fact, Waite et al [10] have shown that as much as 10% of Jupiter’s total incident auroral power goes into vibrational excitation of H₂.

Vibrational levels of ground state H₂, $X^1Σ^+_g (v'' > 0)$, are excited in the atmospheres of Jupiter and Saturn via three predominant mechanisms: (1) direct excitation by electron impact, (2) photon and electron induced fluorescence of the Lyman and Werner bands ($B^1Σ^+_u (v')$ and $C^1Π_u (v')$ – $X^1Σ^+_g (v'')$ respectively), and (3) dissociative recombination of H₃⁺ [7-9]. Once excited, these levels can decay radiatively via the quadrupole interaction or through vibrational-translational and vibrational-vibrational collisions with electrons, ions, atoms, and other H₂ molecules. Majeed et al [9] have calculated the lifetimes of $X^1Σ^+_g (v'' = 1-14)$ in the atmospheres of Jupiter and Saturn where they considered all the above mentioned de-excitation mechanisms. At the altitude of its peak density, the $v''=1$ level has a chemical lifetime of $2.50 \times 10^3$ s and $2.81 \times 10^4$ s on Jupiter and Saturn, respectively [9]. Being so long-lived, vibrationally excited hydrogen molecules have a relatively high probability of interacting with free electrons in the Jovian and Saturnian atmospheres. Such collisions can lead to both emission from excited levels, dissociation of the H₂ molecule, and even de-excitation of the molecule through superelastic collisions.

To date, White and Ross [11] give the only example of energy loss spectra from vibrationally excited hydrogen. In this work, an energy loss spectrum was measured at 0° scattering from 10 to 12.5 eV with a 250 eV electron beam crossed with a fast flowing H₂ beam down stream of a microwave discharge. Calculated Franck-Condon (FC) factors were used to estimate the target $v''=1$ population as 2.5%. However, no cross sections were derived. Given the prevalence of the Lyman and Werner band emissions in the Jovian and Saturnian atmospheres, and the abundance of vibrational quanta present, determining cross sections for excitation of the $B^1Σ^+_u$ and $C^1Π_u$ states out of the $X^1Σ^+_g (v'' > 0)$ levels, is of great importance to outer Solar System aeronomy.

2.1. Experimental

Differential cross sections (DCSs) were measured for the $X^1Σ^+_g (v''=1)$ – $B^1Σ^+_u (v')$ and $C^1Π_u (v')$ excitations via electron energy loss spectroscopy. The electron scattering spectrometer at the Jet Propulsion Laboratory and many of the relevant techniques used in the reported experiments have been described previously [12, 13]. In brief, the apparatus consists of a target beam of vibrationally excited H₂, an energy-selected, well-collimated electron beam source (electron gun), and a scattered-electron detector. Cylindrical electrostatic optics and double hemispherical energy selectors are utilized both in the electron gun and the detector. Energy loss spectra are collected at fixed impact energies and scattering angles by repetitive, multi-channel-scaling techniques. All components are enclosed in a vacuum chamber with double layered magnetic shielding.

2.1.1. A source of vibrationally excited H₂

In order to produce a target of vibrationally excited H₂, a source was designed based on the recombinative desorption method first described by Hall et al [14] and Eenshuistra et al [15]. These works showed that vibrational levels of up to $v''=9$ could be produced by effusing H₂ from a nearly closed metal cell containing a heated filament. In these and subsequent works [16-20], H atoms were produced via dissociation on the hot filament (~2% dissociation), which then recombined on the walls of the cell producing vibrationally excited H₂ via Eley-Rideal reactions, among others. Details
regarding precise mechanisms of vibrationally excited H2 production via recombinative desorption is beyond the scope of this report but can be found in [17] and [20].

The present source, depicted in Figure 1, consisted of a cylindrical stainless steel crucible, which enclosed a Ta filament with an aperture from which the target beam effused. Unlike the experiments cited above, the current source did not rely on the hot filament for dissociation. Instead, we opted to supply the recombination surfaces with H atoms from a microwave discharge. In contrast with the 2% dissociation rate mentioned above, we were able to reliably deliver a mixture of H and H2 from the microwave discharge through a Teflon tube into the recombination cell with dissociation rates of 85% or greater.

Prior to operation of the source, an electric current was passed through the filament in order to sputter Ta from the filament and thereby coat the inner walls of the crucible with a pristine layer of Ta for catalyzed recombination. By depositing the Ta layer in situ and under high vacuum, we avoided/minimized any possible pitfalls that may be associated with oxygen and/or water adsorption to the recombination surface. When operating, the crucible was water cooled in order to promote both Ta adsorption to the steel surface during the conditioning phase and H adsorption on the Ta surface during the experiments.

2.1.2. Analysis and results.

Energy loss spectra were measured at 40 eV impact energy at scattering angles ranging from 7.5° to 60°. Spectra were measured over the energy loss, ΔE, range of ~10 to 12.75 eV. Further spectra covering the elastic region, i.e., from -1.25 to 1.25 eV, were also measured. Figure 2 shows examples of the later with the discharge OFF and ON. From these spectra, we clearly observe a feature that is attributable to the X 1Σg + v”=1 to v”= 0, i.e., X 1Σg + (1-0), superelastic transition at ΔE=-0.5 eV in the ON spectrum versus the OFF spectrum, which is clear of features in the superelastic region. There is no evidence of any other superelastic features in the spectrum. Figure 3 shows examples of discharge ON and OFF inelastic spectra, which clearly indicate the presence of the H(n=2) excitation feature in the discharge ON spectrum, which is not present in the OFF spectrum. We therefore conclude that the only species in the target beam with significant populations are H(n=1) and H2 in the X 1Σg + (v”=0) and X 1Σg + (v”=1) levels.

Spectra were unfolded using a FC based approach similar to that described by [21]. In the measured inelastic energy loss range, it was assumed that only features associated with the B 1Σu + and C 1Πu states were of significance (later verified by quality of fits in the unfolding procedure, see Figure 3). Therefore, potential energy curves and FC factors, calculated via the Rydberg-Klein-Rees (RKR) and direct integration methods, respectively, for the X 1Σg + (0) – B 1Σu + (v’), X 1Σg + (0) –
the \(H(n=2,3,4)\) features and the \(X^1\Sigma_g^+\) factors. Intensities for the remaining \(B^1\Pi_u\) features were held fixed in relation to each other in accordance with the ratios of their FC factors. The intensities of the line-shapes were then varied to produce the

A second fit was then performed. Here, line-shapes were fixed at energy loss values appropriate for the \(H(n=2,3,4)\) features and the \(X^1\Sigma_g^+ (0) – B^1\Sigma_u^+ (\nu^v)\), \(X^1\Sigma_g^+ (0) – C^1\Pi_u (\nu^v)\), \(X^1\Sigma_g^+ (1) – B^1\Sigma_u^+ (\nu^v)\), and \(X^1\Sigma_g^+ (1) – C^1\Pi_u (\nu^v)\) features. The intensities of the line-shapes were then varied to produce the

\[\theta = 7.5^\circ\] with the discharge OFF (left) and ON (right). Fits were made to the indicated inelastic and superelastic features.

Inelastic spectra measured with the discharge OFF showed the onset of the \(X^1\Sigma_g^+ (0) – B^1\Sigma_u^+ (\nu^v)\) manifold occurring at approximately \(\Delta E = 11.2\) eV (see Figure 3). Therefore, the features in the ON spectra, with \(\Delta E > 10.198\) eV (i.e., the \(H(n=2)\) feature) and < 11.2 eV, were attributable to excitation of the \(X^1\Sigma_g^+ (1) – B^1\Sigma_u^+ (\nu^v=0,1,2)\) levels at 10.668, 10.831, and 10.990 eV, respectively. Spectra were fitted by placing the determined line-shape at the energy loss locations corresponding to the \(H(n=2)\) and \(X^1\Sigma_g^+ (1) – B^1\Sigma_u^+ (\nu^v=0,1,2)\) excitation features, where the relative intensities of the \(X^1\Sigma_g^+ (1) – B^1\Sigma_u^+ (\nu^v=0,1,2)\) features were held fixed in relation to each other in accordance with the ratios of their FC factors. Intensities for the remaining \(B^1\Sigma_u^+ (\nu^v)\) levels were then determined, relative to the intensities for the \(B^1\Sigma_u^+ (\nu^v=0,1,2)\) levels obtained in the fit, using the calculated FC factors.

A second fit was then performed. Here, line-shapes were fixed at energy loss values appropriate for the \(H(n=2,3,4)\) features and the \(X^1\Sigma_g^+ (0) – B^1\Sigma_u^+ (\nu^v)\), \(X^1\Sigma_g^+ (0) – C^1\Pi_u (\nu^v)\), \(X^1\Sigma_g^+ (1) – B^1\Sigma_u^+ (\nu^v)\), and \(X^1\Sigma_g^+ (1) – C^1\Pi_u (\nu^v)\) features. The intensities of the line-shapes were then varied to produce the
optimal fit. In the fitting procedure, the relative intensities of features associated within the X $^1\Sigma_g^+$ – B $^1\Sigma_u^+$ and C $^1\Pi_u$ manifolds were held fixed in relation to each other in accordance with their FC factors. Further, the intensities of X $^1\Sigma_g^+$ (1) – B $^1\Sigma_u^+$ manifold were not varied but held fixed at the level determined in the previous step. This procedure produced relative intensities for all atomic lines and vibrational transitions within the molecular manifold of each electronic state.

In order to put the measured relative inelastic intensities on an absolute scale, we relied on an application of the principle of detailed balance and the intensities of the X $^1\Sigma_g^+$ (1-0) and X $^1\Sigma_g^+$ (0-1) transitions, which were determined by fitting these features in consecutively measured elastic spectra.

The principle of detailed balance can be written as,

$$E_0 g_0 DCS_{0-1}(E_0) = [E_0 - \Delta E] g_1 DCS_{1-0}(E_0 - \Delta E),$$

where $E_0$ is the incident electron energy in the inelastic process, $g_0$ is the degeneracy of the initial state in the inelastic process, $\Delta E$ is the inelastic energy loss, $DCS_{0,1}$ is the DCS of the inelastic process, $E_0 - \Delta E$ is the incident electron energy in the superelastic process, $g_1$ is the degeneracy of the initial state in the superelastic process, and $DCS_{1,0}$ is the DCS of the superelastic process. Assuming that the measured intensities, $I$, are directly proportional to the appropriate DCSs and that $DCS_{1,0} (E_0=40 \text{ eV}) \equiv DCS_{1,0} (E_0 - \Delta E=39.5 \text{ eV})$, with the implicit assumption that the transmission function is flat over the energy loss range spanned by the inelastic and superelastic processes, then

$$I_{1-0} = \frac{N_1}{N_0} \frac{DCS_{1-0}}{DCS_{0-1}},$$

where $N$’s give the appropriate target populations of $v''=0$ and $v''=1$ levels. Equations (1) and (2) give

$$I_{1-0} = \frac{N_1}{N_0} \frac{g_1}{g_0} \frac{E_0 - \Delta E}{E_0} \frac{I_{1-0}}{I_{0-1}}.$$  

With the ratio-of-degeneracies equal to unity, the population ratio could be determined, as all other quantities are known. The inelastic DCSs could then be normalized to known inelastic X $^1\Sigma_g^+$ (1) – B $^1\Sigma_u^+ (v')$ cross sections (i.e., DCS$_{0,b}$) [22] via the relation:

$$DCS_{1-X} = \frac{N_0}{N_1} \frac{I_{1-X}}{I_{0-b}} DCS_{0-B}.$$  

The results for the X $^1\Sigma_g^+$ (1) – B $^1\Sigma_u^+ (v')$ and C $^1\Pi_u (v')$ excitations are shown in Figure 4 along with the DCSs for the X $^1\Sigma_g^+$ (0) – B $^1\Sigma_u^+ (v')$ and C $^1\Pi_u (v')$ DCSs of [22] for comparison. Of particular note is the significant enhancement of the C $^1\Pi_u$ DCS for excitation from X $^1\Sigma_g^+$ (1) versus X $^1\Sigma_g^+$ (0) across

![Figure 4](image-url.com)
the measured angular range. Although such enhancement is not seen at low angles in the case of the \( B^1 \Sigma_u^+ \) state excitation, a significant enhancement is seen at 60°. Since the \( \sin \theta \) factor, introduced when deriving integral cross sections, minimizes the low angle contribution, this mid-angle enhancement could prove important in the atmospheres of Jupiter and Saturn. Further work is in progress.

3. Nitrogen

UV spectra of the Solar System’s nitrogen-rich atmospheres all show the importance of excitation of the Rydberg-valence states of \( \text{N}_2 \). In particular, emissions from the \( b^1 \Pi_u \), \( b^1 \Sigma_u^+ \), and \( c^1 \Sigma_u^+ \) states of \( \text{N}_2 \) (i.e., the Birge-Hopfield I and II, and Caroll-Yoshino bands) are prominent in spectra of Titan (Voyager and Cassini), Triton (Voyager), and Earth (FUSE) [4, 23, 24]. Observations such as these provide a strong impetus for study of the energy deposition mechanisms responsible for the excitation and subsequent emission. In terms of the high-lying Rydberg-valence states, experimental and theoretical studies are complicated by strong perturbative mixing between the \( b^1 \Pi_u \), \( c^1 \Pi_u \), \( o^3 \Pi_u \), \( c^1 \Sigma_u^+ \), and \( b^1 \Sigma_u^+ \) states, which has led to significant gaps in our understanding of the relevant excitation processes, predissociation yields, etc.

3.1. Differential cross sections for excitation of the Rydberg-valence states.

Differential cross sections (DCSs) for electron impact excitation of the \( a^\pi \Sigma_u^+ \), \( b^1 \Pi_u \), \( c^1 \Pi_u \), \( o^3 \Pi_u \), \( b^1 \Sigma_u^+ \), \( c^1 \Sigma_u^+ \), \( G^3 \Pi_u \), and \( F^3 \Pi_u \) states in \( \text{N}_2 \) from the \( X^1 \Sigma_g^+ \) (\( v''=0 \)) ground level have recently been measured by our group. DCSs were derived from energy loss spectra covering the energy loss region of 12 eV to 13.82 eV measured at incident energies of 17.5, 20, 30, 50, and 100 eV, and for scattering angles ranging from 2° to 130° [25]. The analysis of the spectra follows a different algorithm from that employed in our previous study of \( \text{N}_2 \) for the valence states [21], since the \( b^1 \Pi_u \) and \( c^1 \Sigma_u^+ \) states form a strongly interacting Rydberg-valence series [26-28], whose consequent quantum interference effects lead to intensity anomalies in vibrational series, which vary with the scattering angle and energy. Similar interactions are expected within the \( ^3 \Pi_u \) manifold [29]. Thus, the invariant FC factor assumption, which is implicit in the spectral unfolding technique of [21] is invalid in this case. The spectral unfolding relied on spectral fitting of all vibrational levels of a given electronic transition treated as independent vibrational transitions. The one exception was the \( a^\pi \Sigma_u^+ \) state, which is unaffected by perturbative interactions with nearby states [30]. Therefore, in its analysis, the intensities of its vibrational-level energy loss features were fixed in proportion to the FC factors for their excitation from the \( X^1 \Sigma_g^+ \) (\( v''=0 \)) ground state.

Figure 5 shows an example energy loss spectrum and its unfolding-fit taken at 50 eV impact energy and 3° scattering angle. In Figure 6, the DCS of the \( a^\pi \Sigma_u^+ \) state is shown as an example. Of particular note is the ‘cusp-like’ feature seen at \( \sim 20^\circ \), which may be attributable to channel-coupling effects, similar to the cusps observed in atomic transitions where the lower and upper states have the same symmetry, e.g., the 1S to 2S transition in He. The discovery of this cusp and its possible interpretation was the subject of a recent paper by our group [30]. Furthermore, integral cross sections (ICSs) for all the transitions investigated by [25] are being derived and will be the subject of a future paper.

3.2. The \( c^1 \Sigma_u^+ – X^1 \Sigma_g^+ \) band system.

Recently, we took part in a collaborative experimental and coupled-channels theoretical study of the emission properties of the \( c^1 \Sigma_u^+ – X^1 \Sigma_g^+ \) band system [31]. Relative intensities of the \( c^1 \Sigma_u^+ (0) – X^1 \Sigma_g^+ (v'' \rangle \) transitions, at the rotational level, were measured via electron impact induced emission spectroscopy and combined with a coupled-channel Schrödinger equation (CSE) model of the \( \text{N}_2 \) molecule. This enabled the calculation of the diabatic electronic transition moment for the \( c^1 \Sigma_u^+ – X^1 \Sigma_g^+ \) system as a function of internuclear distance. A high-resolution, experimental UV emission spectrum was then compared with a model spectrum based on the CSE rotational transition probabilities (see Figure 7). The excellent agreement seen between the measured and modeled CSE spectra provided strong verification of the CSE results.
Reliable calculation of the $c_4^{'} 1\Sigma_u^+ + X 1\Sigma_g^+$ spontaneous transition probabilities, which our results make possible, are important for modelling atmospheric emissions. To illustrate this potential, we point to Figure 8, which shows excellent agreement between the cross section for the $X 1\Sigma_g^+ (0) - c_4^{'} 1\Sigma_u^+ (0)$ excitation derived from our work described above [25] and the results of an emission model based on the CSE transition probabilities [32].

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Figure 5. Energy loss spectrum measured at $E_0=50$ eV and $\theta=3^\circ$ (solid circles–measured data, solid line–the unfolding fit).

Figure 6. DCS for the $a^1\Sigma_g^+$ state at $E_0=30$ eV.

Figure 7. Comparison of measured and modelled $c_4^{'} 1\Sigma_u^+ (0) - X 1\Sigma_g^+ (1)$ band emission ($\Delta \lambda =33$ mÅ, $E_0=100$ eV, $T=260$ K); see [31].

Figure 8. Comparison of cross sections for the $X 1\Sigma_g^+ (0) - c_4^{'} 1\Sigma_u^+ (0)$ excitation. Model cross section (line) is rotationally averaged at 300 K.
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