Optical emission produced by proton and hydrogen atom impact on nitrogen and hydrogen molecules
by Duane Arlen Dahlberg

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Abstract:
Optical emissions produced in collisions of protons incident on nitrogen and hydrogen molecules and hydrogen atoms incident on the same molecules were studied in the spectral region from 1200 Å to 6000 Å. Relative emission cross sections were measured in the energy range from 10 kev to 130 kev.

The spectra excited by proton impact with nitrogen and hydrogen molecules were scanned with a monochromator and photomultiplier. The prominent features of the nitrogen spectrum below 2000 Å were the Lyman alpha line and some atomic nitrogen lines. The Lyman-Birge-Hopfield system appeared also, but it was weak. At the longer wavelengths the N_2^+ First Negative and the N_2 Second Positive systems dominated the spectrum. The Balmer beta and an ionic nitrogen line appeared as well. Both Lyman alpha and the Lyman bands were prominent in the hydrogen molecular spectrum. Lyman alpha, however, was the dominant emission. In addition to the Balmer lines, a few lines of the hydrogen molecular spectrum were present in the longer wavelength spectral region.

Relative emission cross sections were measured for the production of a number of the First Negative bands, the (0,0) Second Positive band, an ionic nitrogen line, atomic nitrogen lines, and Lyman alpha in proton and hydrogen atom collisions with nitrogen gas. Cross sections for the production of the First Negative bands in collisions of hydrogen atoms with nitrogen molecules were nearly constant as a function of energy, and at an energy of 40 kev the cross section was one-half as large as the cross section for proton collisions. The cross section for the Second Positive band due to hydrogen atom impact was about 3×10^{-18} cm^2 at 25 kev whereas for proton impact the cross section was about 2×10^{-19} cm^2 at its maximum value. The cross sections for the atomic nitrogen lines produced in hydrogen atom impact were approximately 75% of the cross sections for the same lines produced in proton impact. Hydrogen atom collisions had higher cross sections throughout the energy range for the production of Lyman alpha emission.

Relative emission cross sections for the production of the Lyman bands and Lyman alpha by proton and hydrogen atom impact on hydrogen molecules were also measured. Hydrogen atoms proved to be much less effective than protons in exciting the Lyman bands of hydrogen.

Consideration was given to the possible mechanisms responsible for the various emissions and comparisons were made to the energy dependence predicted in the Born approximation theory.

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CHAPTER I. INTRODUCTION

The homonuclear diatomic molecule represents the simplest molecular structure and the proton is the simplest atomic nucleus. Collisions between these systems would be the least difficult of all ion-molecule collisions to analyze, and the experimental data obtained in studying these collisions would be most useful in evaluating approximate theoretical treatments. Since the hydrogen atom is the simplest atomic structure, collisions between the homonuclear diatomic molecule and the hydrogen atom represent the least complex of the atom-molecule interactions. The hydrogen atom differs from the proton only by the bound electron of the hydrogen atom. Experimental data obtained for collisions in which protons are replaced by hydrogen atoms, therefore, are also of theoretical interest. The least complex of the diatomic molecules is the hydrogen molecule, and consequently, collisions involving this molecule are of the greatest theoretical interest.

One method for studying collisions between two systems is to measure the photon energy which is emitted in optical transitions resulting from the collision. For particular cases a measurement of the photon emission provides a direct means of obtaining cross sections for exciting atomic and molecular states. A comparison of these experimentally obtained excitation cross sections with theoretical predictions can assist in understanding collision mechanisms.

Atmospheric research indicates that fast protons are entering the atmosphere and contribute to the production of auroras[1]. Because of the charge exchange process whereby protons pick up electrons from atmospheric gases, there would also be fast hydrogen atoms present.

It is evident from the work of Allison and others[2] that the equilibrium fraction of hydrogen atoms produced in charge exchange is significant for projectile energies of a few kev to 100 kev. It is of interest, therefore, to determine the effectiveness of the hydrogen atoms in exciting the atmospheric gases and also their contribution to the auroras. The knowledge of hydrogen atom excitation cross sections is also important in studying the history of a proton entering the atmosphere.

For these reasons optical emissions produced in collisions of protons and hydrogen atoms with nitrogen and hydrogen molecules have been studied.

The experimental apparatus used to measure the light emitted in both proton and hydrogen atom collisions represents an integral part of this work and will be discussed next. The proton beam from a Cockroft-Walton accelerator equipped with an RF ion source is focused and mass analyzed. The resulting beam passes through a set of collimating apertures in the charge exchange cell, then into a differentially pumped region, and finally into the excitation chamber. Finally the protons are collected in a Faraday cage at the end of the excitation chamber. If a hydrogen atom beam is desired, a molecular gas is admitted to the charge exchange cell. In passing through the charge exchange cell the initial proton beam becomes a mixture of hydrogen atoms and protons. The protons are electrostatically deflected from the beam in the differentially pumped region, leaving only the hydrogen atoms to enter the excitation chamber. The hydrogen atom beam current is measured by secondary emission techniques. The light which is emitted in the collisions between the incident particle beam and the target gas is chopped mechanically and spectrally analyzed with a monochromator. A photomultiplier and a phase sensitive detection system are used to measure the light intensity.

Spectral scans of the light emitted in the collisions of protons with both nitrogen gas and hydrogen gas in the spectral region from 1200 Å to 6000 Å indicated which transitions of the molecules, of the atoms and ions originating from molecular dissociation, and of the incident particle produced sufficient light intensities for emission cross section measurements. The results of the scans led to relative emission cross section measurements for the production of the N_2^+ First Negative band system ($B\ ^2\Sigma_u^+ --- X\ ^2\Sigma_g^+$), the N_2 Second Positive band system ($C^3\Pi_u --- B^3\Pi_g$), atomic and ionic nitrogen lines, Lyman bands in hydrogen, and the Lyman alpha line. The emission cross sections for these molecular vibrational bands and atomic lines were measured as a function of projectile energy from 10 kev to 130 kev. Both protons and hydrogen atoms were used as the incident particles.

Optical emissions produced by protons incident on nitrogen have been studied in the visible and near ultra-violet spectral region by a number of researchers [3,4,5,6]. Some emission cross section measurements of Lyman alpha produced in proton collisions with nitrogen and hydrogen molecules have also been reported [7,8]. Since no emission cross sections for hydrogen atoms incident on nitrogen and hydrogen molecules have been measured, a particular emphasis was placed on the emissions due to hydrogen atom impact. In the vacuum ultra-violet spectral region no emission cross sections for proton and hydrogen atom impact had been measured for these molecules. An effort was made, therefore, to also measure emission cross sections for optical transitions in the spectral region from 1200 Å to 3000 Å.

In those cases where emission cross sections could be directly related to the excitation cross sections, the measured cross sections were qualitatively related to theory.

CHAPTER II. THEORY

The primary purpose of this discussion is to present briefly some of the most pertinent aspects of the theoretical work involving ion-atom and atom-atom collisions. Some of these theoretical results can be helpful in understanding ion-molecule and atom-molecule collisions. The main body of theoretical work has been centered in collisions of electrons or ions incident on atoms. Discussions of collision processes, collision cross section, some basic theoretical treatments, and a few results are included in this chapter.

COLLISION PROCESSES

There are two basically different processes which occur in a collision. One is the elastic collision in which the kinetic energy is conserved. The second is the inelastic collision in which the kinetic energy is not conserved.

Elastic scattering produces no effect other than changes in kinetic energy and momentum of the colliding particles.

For the inelastic collision the situation is very complex. If an electron is the accelerated particle and A is an atom, the following basic reactions are possible:

$e + A \text{ --- } e + A^*$	Excitation
$e + A \text{ --- } e + A^+ + e$	Ionization
$e + A_2 \text{ --- } e + A + A$	Dissociation

If an atomic ion or atom is the accelerated particle, the additional reactions can occur:

$B^+ + A \text{ --- } B + A^+$	Charge Exchange or Electron Capture

$B + A \text{ --- } B^{+} + A + e$ Stripping

$B^{*} + A \text{ --- } B + A^{*}$ Excitation Transfer

In addition many combinations of these basic processes are possible in a collision. Some examples that will be considered in this paper are charge exchange plus excitation, ionization plus excitation, and dissociation plus ionization plus excitation. For a more complete listing and classification of collision processes the reader is referred to Hasted's work [9].

COLLISION CROSS SECTION

Both elastic and inelastic collision probabilities are defined in terms of the cross section. The total collision cross section, Q_t, can be expressed as $\sum_i Q_i$, where Q_i represents the cross section for an individual type of collision process and the sum is over all possible collision processes.

For two hard spheres of radii r and r' the cross section for elastic collision is defined simply as $Q_e = \pi (r + r')^2$. For inelastic collisions involving ions and atoms the expression for cross section is complex due to the large number of individual processes contributing to the cross sections.

Although many processes are indirectly connected to the work discussed in this paper, the process of direct interest is the excitation process. An expression can be obtained which relates the emission measurements to excitation cross sections.

Consider the excitation produced when a beam of particles passes through a chamber containing a target gas. The change in the number of excited target atoms in state "i" per second over a path length of one cm, as a consequence of collision with beam particles, is expressed as

$$(dn_i/ dt) = NIQ_i + \sum_{k>i} A_{ki}n_k - \sum_{j<i} A_{ij}n_i$$

In this equation n_i is the number of atoms excited to state "i" per cm path length; N is the number of target atoms per cm^3; I is the number of particles passing through a unit surface perpendicular to the beam direction; and A_{ij} is the transition probability from state "i" to "j". Q_i is the cross section for excitation of level "i". The first term on the right in this equation represents the direct excitation to state "i"; the second term is the population by cascading from higher levels, and the third term represents the decay of state "i" to all lower levels. At equilibrium $(dn_i/dt) = 0$. If one neglects the cascading term $Q_i = \sum_{j<i} A_{ij}n_i = \sum_{j<i} Q_{ij}$, where Q_{ij} is the effective cross section for emission of the line representing the transition from "i" to "j". If cascading is included, $Q_i = \sum_{j<i} A_{ij}n_i - \sum_{k>i} A_{ki}n_k$.

The cross section for the excitation of the fast particle consists of the same terms as for the target particle, providing that equilibrium has been established between the formation of the excited state and emission. Absolute measurements, however, require a calculation of the effective path length of observation.

Cross sections normally are expressed in terms of cm^2/ target particle.

THEORETICAL TREATMENTS

Classical Method of Gryzinski

Briefly, Gryzinski's approach[10,11,12] to atomic collisions is based upon the assumption that the interaction between a charged projectile and an atom can be described classically by the coulomb interaction between the projectile and the atomic electrons. This is a classical theory, but its results depend upon the binding energy and the momentum distribution of the atomic electrons which are obtained quantum mechanically. The Gryzinski theory has been used for calculating cross sections for some inelastic collisions, but calculations for excitation processes have not been made. The moderate success of the theory for some inelastic collisions[12] should provide the impetus, however, for excitation calculations.

Quantum Mechanical Methods

Since classical methods are generally not able to predict the behavior of inelastic collisions, quantum theories must be used.

There are two different approaches to the theory which have been exploited. These are the impact parameter treatment and the wave treatment[13]. The impact parameter treatment is applied in particular to collisions of heavy particles. The assumptions are made that the nuclei can be regarded as classical particles and that the nuclei follow classical trajectories which approach straight lines as the particle velocity increases. This treatment is semiclassical since quantum mechanics is used for describing the electron motion. The wave treatment assumes that the incoming and outgoing particles can be treated as waves. This treatment can, therefore, be used as effectively for electrons as for heavy particles. For collisions involving heavy particles, however, these two treatments have been shown to be mathematically equivalent [14].

In quantum mechanics the motion relative to the center of mass of a system of two colliding particles can be described by the following wave equation [15]:

$$\left[(\hbar^2/2M)\,\nabla_r^2 - H_a(\vec{r}_a) - H_b(\vec{r}_b) + \tfrac{1}{2}Mv^2 + E_o - V(\vec{r},\vec{r}_a,\vec{r}_b)\right]\Psi = 0$$

In order to analyze this wave equation, each section will be considered separately.

First of all, the motion of one particle relative to the other can be represented by the equation:

$\left[(\hbar^2/2M)\,\nabla_r^2 + \tfrac{1}{2}Mv^2\right] F(\vec{r}) = 0$, where $\vec{r}$ and v are, respectively, the relative coordinates and velocity of the two particles and M is the reduced mass.

Second, the internal motion of the two particles can be represented by the equation: $\left[H_a(\vec{r}_a) + H_b(\vec{r}_b) - E_a - E_b\right]\Psi = 0$, where $\psi(\vec{r}_a,\vec{r}_b)$ is some product of $u(\vec{r}_a)$ and $v(\vec{r}_b)$. The equations, $\left[H_a(\vec{r}_a) - E_a\right] u(\vec{r}_a) = 0$ and $\left[H_b(\vec{r}_b) - E_b\right] v(\vec{r}_b) = 0$ describe the internal motion of particle a and particle b, respectively, where H_a and H_b are respectively the Hamiltonians for particles a and b.

Third is the term which prescribes the interaction between the two particles, $V(\vec{r},\vec{r}_a,\vec{r}_b)$.

In principle the wave equation describing a colliding system can be solved and the cross sections for particular scattering events can be obtained. In practice, however, values for cross sections can be calculated only by approximate means, even for the simplest system.

The two quantum mechanical approximations which have application here are the Born approximation and the distorted wave approximation.

Born Approximation: The wave treatment of the Born approximation is discussed extensively by Mott and Massey [15] and the impact parameter treatment is considered in detail by Bates [13].

Basically the Born approximation assumes that the incident particle is an undistorted plane wave which is unaffected by the interaction with the target. The interaction producing the excitation is assumed to be small so that the effect on the incident wave can be neglected. Any excitation that occurs results from the direct excitation from the initial to the final state. The coupling between intermediate states is, therefore, ignored.

These assumptions imply that the approximation is valid only at high impact velocities. The term "high" must be expressed more definitely. The only means by which this can truly be accomplished is to compare the theoretical results to experiment. In general terms, however, the velocity is considered high if $(e^2/hv) \ll 1$, where v is the velocity of the incident particle, and if $v^2/u^2 \gg 1$, where u is the orbital velocity of the atomic electrons. In terms of momentum the velocity is high if $K_o^2 \gg K^2$, where $K_o^2 = 2m\,|E_o|/\hbar^2$, and E_o is the excitation energy of the target particle. $K\hbar$ is the change in momentum of the incident particle. For example, in the ionization of helium by proton impact, the Born approximation is reasonably good above 60 kev [16]. For further discussion of this subject refer to the work of Bates [13].

In the Born approximation equations for cross section have been formulated also in terms of the energy of the incident particle and transition probabilities [15]. This formulation is sometimes referred to the Bethe-Born approximation and is significant because it provides equations relating the energy dependence of the cross sections for ionization, excitation, and charge exchange produced in collisions of fast ions with atomic systems. The cross section for optical excitation of an allowed transition or outer shell ionization of an atom by a fast ion is:

$$Q_{nl,n'l'} \approx (\text{Constant}/mv^2) \left| x_{nl,n'l'} \right|^2 \ln(2mv^2/E_{n'l'}-E_{nl})$$

$\left| x_{nl,n'l'} \right|^2$ is the square of the dipole transition probability. $E_{n'l'}$ and E_{nl} are energies of the n'l' and nl levels, respectively. If excitation is to a forbidden level and the transition is associated with the quadupole moment, the cross section will have the form:

$$Q_{nl,n'l'} \approx (\text{constant}/v^2) \left| (x^2)_{nl,n'l'} \right|^2 E_{nl}$$ [15].

<u>Distorted Wave Approximation:</u> Mott and Massey have also described the distorted wave approximation and its semiclassical counterpart, the distortion approximation [15]. The two different names are connected, respectively, with the wave treatment and the impact parameter treatment of this approximation.

As in the Born approximation one assumes that the coupling between the two states under consideration is weak. Instead of assuming plane incoming and outgoing waves, as is done in the Born approximation, the distorted wave approximation allows for a distortion of the incoming and outgoing waves by the scattering potential field. The transition matrix elements, therefore, become a function of the interaction potentials. The distorted wave approximation gives a lower value of cross section than the Born approximation at low energies. At high energies the two approximations approach the same values and are mathematically equivalent.

RESULTS OF THE THEORY

In this section the applications of the theory, comparisons of the Born and distortion approximation, and the general dependence on energy will be discussed.

Applications and Comparisons

In 1953 Bates published a discussion and critique of the various approximations used for describing the inelastic collisions between ions and atoms [17]. At that time the Born approximation was applied at the higher energies and at the lower energies the perturbed stationary state approximation was used. In a more recent publication [18] Bates questioned the validity of ignoring the effect of the scattering field on the incoming and outgoing waves. This led to the distortion approximation for calculating inelastic cross sections for ion-atom collisions. Since then a number of researchers have calculated cross sections for the excitation of hydrogen and helium by proton impact. They have used either the Born or Distortion approximations.

Using both the Born and Distortion approximations Bell [18] has calculated the cross sections for the formation of the $3\,^1P$ excited state of neutral helium by proton impact. These calculations have been compared with experimental results by J. Van Eck, et. al. [20] and E. W. Thomas, et. al. [21].

Bates and Crothers [22] have gone a step further. They have considered atom-atom collisions and have taken into account the exchange of electrons between the incident particle and the target atom. This work was then applied specifically to the excitation produced in the collision of hydrogen atoms on helium.

The exchange of electrons is probably the dominant mechanism in the excitation of a triplet state from a singlet ground state. The calculations were, therefore, compared with the cross sections for the excitation of the $3\ ^3P$ state of helium measured by J. Van Eck, et. al. [20]. The comparison was quite good for this triplet state but not so good for some of the other triplet states of helium.

General Energy Dependence of Cross Section

At high incident particle energies the cross sections for inelastic collision processes have a dependence on energy described by the Born approximation. For simple excitation and ionization Q is proportional to (1/E) ln E. For electron capture [9] Q is proportional to E^{-6}. At the high incident particle energies the cross section plotted as a function of energy will, therefore, decrease as the energy increases.

For low energies the cross sections for inelastic collisions will decrease with decreasing energy. Consequently a maximum in the cross section must exist at some intermediate energy. The adiabatic criterion proposed by Massey [23] is useful in predicting the energy dependence at low energies and in predicting the maximum in the electron capture process. At low impact energies where the velocity of relative motion is small, the electron is able to adjust to changes in internuclear distances. The electron, therefore, has a small probability of making a transition and consequently the cross section for the transition is small. As the relative velocity, v, increases the probability for transition of an electron increases and the time of collision, a/v, decreases. The adiabatic parameter, "a", is the distance in which the electron capture process is assumed to be possible. The assumption is made that the cross section for electron capture has a maximum when the time of collision is equal to the time of transition, $\Delta E/h$, where ΔE is the internal energy defect resulting in the electron capture. For the process $A^{+} + B \dashrightarrow A + B^{+}$, the energy defect is defined as $E_B - E_A$, where E_B and E_A are, respectively the ionization energies of atom B and atom A.

This entire argument can now be reversed. If the incident particle velocity at the maximum in the electron capture cross section can be experimentally measured, the Massey criterion provides a method for determining the interaction distance, "a". The determination of "a" has been made for many colliding systems. The interesting result is that the value of "a" is approximately equal to 7 Å for most electron capture processes without regard for the particular atoms involved [24].

Hasted [9] points out that there is sufficient justification for applying the adiabatic criterion to other processes also. According to Solov'ev [25] the application of the Massey criterion to ionization process gives values of "a" ranging from 4.7 Å to 6.6 Å.

Hasted [9] also cautions one in using this criterion for collision processes between more complex systems. An example of the necessary caution is presented in the work of J. Van Eck, et. al.[20]. Applying the criterion to atom-atom collisions which result in excitation of the triplet states of helium, the interaction distance is about 3 Å and this small value of "a" is thought to be due to the formation of intermediate molecular states. If intermediate states are formed, ΔE probably changes in the interaction and the adiabatic criterion can no longer be used.

CHAPTER III. EXPERIMENTAL APPARATUS

The various pieces of experimental apparatus used in the measurements of the emission cross sections produced in collision processes will be discussed. Figures 1 and 2 show the sections of the apparatus which are of special interest in these measurements.

ACCELERATOR

A Cockroft-Walton accelerator manufactured by the Texas Nuclear Corporation (Model 9501-1, Ser. No. 42, Neutron Generator) was modified to provide a stable proton beam of energies from 10 kev to 130 kev. The protons were produced in an Oak Ridge type RF ion source[26] theoretically capable of a total beam of 1 ma, 90% of which would be protons.

A standard resistor equal to $1/10^5$ of the measured resistance of the accelerator tube resistor chain was inserted at the low potential end of the chain. This chain of 20 10 meg Ω resistors acted as a potential divider for the accelerating region. The potential across the standard resistor was directly related to the potential across the accelerator tube and was used to measure the proton energy. From direct measurements at low potentials and comparisons with the readings of a voltmeter for the high voltage supply, the author concluded that the error in energy measurements was never more than 10%.

In preparing this machine for use the following modifications were made:

1. The diffusion pump electrical power system was redesigned in order to provide safety switches and controllable power to the heater of the diffusion pump. Figure 3 shows the electrical circuit. This circuit was used also for the vacuum system on the differentially pumped

chamber.

2. The vacuum plumbing was changed to permit roughing the system while the diffusion pump was on.

3. The cold cathode type pressure gauge circuit was removed from the accelerator control panel. A VEECO ionization gauge was used to monitor the pressure in the accelerator tube and served also as a high pressure safety control.

4. The solenoid field coil on the ion source was replaced with a new design which had a heat sink. The control for varying the solenoid field was moved from the high potential end to the low potential end of the accelerator. This modification provided a means of adjusting the magnetic field in the ion source while the accelerator was in operation.

5. The resistor chain on the accelerator tube was replaced after finding the existing resistors unreliable. These faulty resistors led to beam instabilities.

6. Due to fluctuations in line voltage a Sola regulating transformer was installed to control the power input to the high voltage power supply. This regulating transformer also stabilized the power to all the circuits in the high voltage section of the accelerator.

ELECTROSTATIC QUADRUPOLE LENS

To supplement the focusing already built into the accelerator an electrostatic quadrupole strong focusing lens (Texas Nuclear, Model 9515) was added between the accelerator and the analyzing magnet. A beam could then be maintained even at low energies. This lens also served to correct any aberrations produced by the analyzing magnet.