CHARGE EXCHANGE IN THIN FOILS

by

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The work for this thesis was carried out entirely by the candidate, in the Physics Department of the Australian National University, under the supervision of Professor D.N.F. Dunbar.

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Figure 1
INTRODUCTION

The proton, as well as existing as a single particle, can have one or two electrons bound to it. With one electron we have the hydrogen atom and with two, the negative hydrogen ion. Considering the bare proton as a positive hydrogen ion, we talk of the three charge states of hydrogen: positive, negative and neutral.

If a number of hydrogen particles are passing through matter, each particle will be continually changing its charge through the capture and loss of electrons. After the particles have passed through a certain quantity of matter, an equilibrium concentration of the three charge states will be established, when each type of particle is created and destroyed at an equal rate. At equilibrium, the relative abundance of the three charge states depends on the velocity of the protons and the nature of the matter through which they are travelling.

There has been a lot of work done on the variation of the equilibrium charge ratios as a function of the atomic number of the target medium. On the other hand, very little thought has been devoted to how the ratio might be influenced by the phase and crystal structure of the medium, and on whether it is a metal or dielectric. This is despite the fact that it has been found that a quantity related to the charge ratios, the stopping power, is affected by the chemical state, phase and the orientation of the crystal planes of the stopping medium.
The purpose of performing charge exchange experiments, whether in gaseous or solid matter, is to arrive at an understanding of the mechanisms involved in the electron transfer processes and in atomic collision phenomena generally. Also knowledge of the equilibrium charge ratios is necessary for the theory of stopping, in which the concept of equivalent charge is used to explain the reduced stopping power of matter for low energy ions. On the practical side, data on charge ratios is required for the design of machines, such as the Tandem electrostatic accelerator, in which the charge of the accelerated particles is changed.

Although a great deal of theoretical and experimental work has been carried out on charge exchange collisions in gaseous matter, there has been very little thorough investigation of the phenomenon in solid matter. The aim of the investigation described in this thesis was to measure the relative numbers of particles in the three charge states of hydrogen, after equilibration at various energies in several different solids. An equally important object was an investigation of the many factors which might influence the equilibrium charge ratios.

To measure the equilibrium charge ratios characteristic of solids, it is most convenient to have the solids in the form of thin foils, which are produced by the only suitable technique available, vacuum evaporation. The factors which could affect the charge ratios of the emergent
particles were all associated with the surface and structure of the foils. The importance of minimizing the adsorption of foreign matter on to the surface of the foil had already been realized. However, investigators in this field have paid no attention to the variation in structure of films produced by vacuum evaporation. Far from being the continuous laminar structures often visualized, evaporated films nearly always consist of aggregations of crystallites, contaminated by embedded gas molecules and are often punctured by pinholes.

The basis of the present work was the measurement of the charge ratios of hydrogen particles emergent from thin metal foils. Apparatus was designed and constructed to produce a beam of accelerated protons, which were passed through a thin foil evaporated in situ under good vacuum conditions. Artificial zeolite, used in a special cold trap, ensured a low gas pressure in the vicinity of the foil. A particle analysing and detecting system completed the equipment, which was then used to measure the charge ratios characteristic of aluminium, copper, silver and gold. The study included an investigation of the effect of gas adsorption and attention was also paid to the physics of the condensation of thin metal films in vacuo.

The investigation proved to be useful, for the previously measured proportions of negative ions were found to be too low. Also a hitherto neglected experimental factor, the aggregated structure of evaporated films, was shown to exert a considerable influence on the results. An analysis
of the results leads to the hypothesis that the charge exchange processes are very similar in solids and gases, except that the greater electron density in solids increases the probability of a proton capturing two electrons in a single collision.
Interest was first aroused in charge exchange phenomena, when, about 1890, beams of fast particles, which could not be deflected by electric or magnetic fields, were observed in the canal rays of discharge tubes. The high speed of these particles could only be explained by the fact that they had been accelerated while in charged states. It was realised that the fast ions in the discharges were subsequently neutralized in charge exchange collisions with gas molecules in the discharge tube.

Interest was further stimulated by observations of charge exchange in alpha particles emitted from radioactive materials, but for many years very little work was done on the subject, nuclear physics claiming most attention. More recently there has been a revival of interest, in many cases stimulated by controlled thermo-nuclear research and investigations of the earth's upper atmosphere.

Charge exchange experiments, whether with a target in the form of a gas, a foil or a solid surface, are designed to increase our knowledge of the charge exchange processes. Investigations in the particular field of charge transfer in thin foils lead us to speculate on the following questions:

(a) How closely does the process in foils resemble that in gases?
(b) What effect, if any, do reactions between the emerging particles and the foil surface have on the measured charge ratios?

(c) How important to the charge exchange process is the crystal lattice structure of the evaporated films?

(d) How reliable are measurements of the equilibrium ratios? Alternatively, to what extent do the results depend on factors such as the vacuum conditions or evaporation technique of a particular laboratory?

As a background to later discussion of these questions a brief review will be made of:-

A. The theoretical and experimental work on charge exchange collisions with gaseous targets.

B. Experiments on surface interactions that could be relevant, and theories related to them.

C. Previous experiments on charge exchange in thin foils.

A. CHARGE EXCHANGE IN GASES

Many early experiments were carried out by Henderson and Rutherford with alpha particles from radioactive decay (43, 44, 79). They observed fast singly charged helium ions emerging from radioactive substances, which they attributed to the fact that alpha particles picked up electrons on the way through the emitter. They were also able to measure the capture length for the formation of He from He in air. The energies of the particles were high and so the
theories which were developed also dealt with energetic alpha particles. In general, these theories were taken as being equally applicable to solid and to gaseous targets and at the high energies involved this was probably justified.

One of the earliest of these theories was published by Thomas (88), who attempted to calculate the probability of electron capture by classical means, treating the three body problem by splitting it into a pair of two body collisions. The incident particle was assumed to accelerate an electron in a close collision. This electron was then deviated in the same direction as the incident particle by the nucleus of the target atom. Thomas was able to calculate the capture probability, using approximations which were justified in the case of light target atoms and a limited energy range. In a second calculation he made use of the Hartree results for atomic fields. The approximations he made were justified for an increased velocity range, provided the target atom was heavy. The expression obtained showed the proportion of singly to doubly charged particles to vary as \( V^{-4} \) and not to depend greatly on the nature of the target nucleus.

Shortly afterwards, Oppenheimer (71) published a quantum-mechanical calculation of the capture rate of electrons by alpha particles. He applied his quantum theory of aperiodic effects (70) to a collision involving an alpha particle, an atomic nucleus and an electron. In effect, this
Figure 2

Cross section for charge transfer in collisions between protons and hydrogen atoms.

\( p + H \rightarrow H + p \)

- Experimental data
- Dalgarno and Yadav
- Bates and Dalgarino
- Jackson and Schiff
- Brinkman and Kramers
- Bassel and Gerjuoy

\( \sigma \) (\( \text{cm}^2 \)) as a function of ion energy (keV).
calculation was equivalent to a first Born approximation. He also obtained a $V^{-4}$ dependence for the proportion of electron-carrying to normal alpha particles.

The Oppenheimer theory was developed by Brinkman and Kramers (16). Like his, theirs was a first Born approximation, but was carried through more rigorously. They simplified the interaction potential by including only the interaction between the electron and the two heavy particles and ignoring the interaction between these two heavy particles. The neglect of the heavy particle interaction they justified by pointing out that this interaction would merely produce a deviation of the particles, leaving the electron unaffected. Applying the theory to the calculation of the cross section for electron capture by alpha particles in nitrogen, they compared their results with the experimental values of Rutherford (79). By choice of suitable parameters they were able to obtain an agreement to within a few percent. Their results applied to hydrogen particles are shown in Figure 2.

Up to 1930 all work had been carried out with alpha particles. The first measurements with hydrogen ions were made by Bartels (9), who passed a beam of protons through air, oxygen, nitrogen and hydrogen at various pressures and measured the ratio of positive to neutral particles emergent. He observed that this ratio depended on the proton energy, the nature of the gas and its "thickness". The "thickness" was given by the product of the length of the collision cell and the gas pressure in it. Once the gas exceeded a certain
Figure 3

Electron-capture cross section $\sigma_r$ per atom for protons in hydrogen gas as a function of incident proton energy. The solid curve labeled $\sigma_r$ is the theoretical result [Eq. (19)] for the total capture cross section; the dashed curve labeled $\sigma_1$ is the partial cross section for capture into the ground state [Eq. (17)], while the dotted curve labeled $\sigma_{BK}$ is the corresponding cross section due to Brinkman and Kramers [Eq. (18)]. The experimental data are those of Keene (●), Ribe (■), and Whittier (○).
thickness, the ratio of positive to neutral particles attained a constant equilibrium value. Bartels measured this equilibrium ratio between 15 and 45 KeV. This energy is in the range of interest to the present investigation, but more recent measurements of the equilibrium ratio in gases are used in Chapter 5, when comparison is made with the ratios from foils.

The subsequent extensive experimental work on charge changing collisions in gases has been reviewed by Allison and Warshaw up to 1955 (1,2) and later by Bates (11). A review of the theoretical work has been made by Gerjuoy (35).

Theoretical studies of the phenomenon were resumed in 1953, when several workers criticised the omission of the heavy particle interaction from the Born approximation (12, 46). They recalculated the case of protons travelling through atomic hydrogen, with the proton – proton interaction included. By assuming that a hydrogen molecule is equivalent to two hydrogen atoms, they were able to compare their calculated values with the experimental results for protons in molecular hydrogen. The agreement was good and this justified the inclusion of the proton – proton interaction in the calculation (Figure 3).

However, the calculated cross sections can now be compared directly with measured values in atomic hydrogen (14, 27, 28). The experiments with atomic hydrogen are the most recent in the field of charge exchange and present many experimental difficulties. The target of hydrogen is produced
as an atomic beam from an oven. But, as the concentration of hydrogen atoms in the collision region is low, the beam has to be modulated mechanically to distinguish the interactions due to the hydrogen from those due to the residual gas in the vacuum system.

The experimental results for the electron capture cross section of protons in atomic and molecular hydrogen are shown in Figure 2. Inserted for comparison are the theoretical results discussed above. Also included are the results of the perturbed stationary state approximation of Dalgano and Yadav (22), which are in good agreement with the atomic hydrogen experiments for energies below 10 KeV. It can be seen that the cross section in molecular hydrogen is not twice that in atomic hydrogen.

These results support theoretical objections raised by Tuan and Gerjuoy (90) to the treatment of the hydrogen molecule as two atoms. In a more rigorous calculation, Bassel and Gerjuoy (10) found that the proton-proton interaction was counteracted by another term and that these cancelled at energies above several hundred KeV, to yield the Brinkman and Kramers result. Rigorous calculations of charge exchange collisions have also been made by Mittleman (61, 62).

The fact that charge exchange cross sections are modified by the states of the electrons in molecules is of significance for experiments with metals. For, when metallic atoms condense to form crystals, their electronic structure is modified in a way similar to that of atoms in
the formation of molecules.

It is also of interest to note that there is evidence for some dependence of stopping power on the chemical state of the stopping medium (15, 48).

B. SURFACE INTERACTIONS

In his work on foils, Phillips (74) suggested that "the mechanism proposed by Oliphant and Moon et al. for the ionization and neutralization of the helium atom in the field of the surface of a metal may be applicable to protons."

In this statement Phillips implied that the main problem, in applying the theory of Oliphant and Moon to charge exchange in foils, is in the change from helium to hydrogen. In fact this is not the case, the difficulty lying in the different energy ranges of the two processes. The energy range of the surface interaction work was from 0 to 1000 eV, whereas the lower energy limit for the foil experiment is 5 KeV. Despite this, the investigations of Oliphant and Moon, and also of others in the field of interactions at surfaces, are worth considering for their relevance to charge exchange in foils.

It was during his research on secondary electron emission in 1929, that Oliphant (67) observed that helium ions could be neutralised in glancing collisions with metal surfaces. The atoms formed by the neutralisation retained a high proportion of the energy of the ion and were largely in a metastable excited state. The metastable atoms were detected by means of the secondary electron emission they
Figure 4

Energy

i/e+ (per cent)

Cold

Hot

Figure 4
caused when incident upon a metallic surface. The neutral atoms also caused sputtering of the metal surface, but not as much as positive ions.

In some later work, Oliphant (68) bombarded cold gas-covered and hot gas-free metal targets with He$^+$ ions. He measured the secondary electron yield, and also the velocity distribution of the ejected electrons, as functions of the incident ion energy, which varied between 0 and 1000 electron volts. Figure 4 illustrates the sort of result obtained. It can be seen that the gas completely obscures the threshold for sudden increase in secondary yield, which was observed with hot surfaces.

In a paper published at the same time, Oliphant and Moon (69) examined, in a qualitative way, various secondary emission theories. They considered the formation of neutral atoms by auto-electronic emission. In this process, an electron is extracted from the metal by the electric field of the ion and is later bound to the ion. The probability of neutral atoms being formed by this means was shown to be high when the ion was two or three atom diameters from the metallic surface. Hence they concluded that most slow ions were neutralised before striking the surface.

Another hypothesis they discussed was the neutralisation of ions as an electron transition between states of equal energy. Here the metal provides a source of electrons with a continuous energy distribution, and the energy taken to extract the electron from the metal is equal
Comparison of Calculated and Observed Velocity Distributions.

I, Experimental; II, Theoretical for large distances from the surface; III, Theoretical for zero surface distance.
Calculations along these lines were carried out by Massey (56), who applied the quantum theory giving the probability of transition from discrete states to a continuous set of states, by using it in reverse. In this case the transition takes place between the continuous metallic states and the discrete bound states of the free atom. Massey calculated the velocity distribution of the secondary electrons ejected by positive ions, by considering the interaction as taking place in two stages. First the ion extracts an electron as already indicated. The neutral atom so formed gives its excess energy to a second electron, which is then energetic enough to escape the work function barrier, as in the process of Auger ejection. This calculation was later refined mathematically (57) to give the results shown in Figure 5. Two results are given; in one case the distance between the ion and the metal surface is taken to be large at the time of interaction, and in the other it is taken to be small. These results do not differ greatly and can be seen to agree well with Cliphant's experimental value of velocity distribution.

A similar calculation was carried out by Smith (83) to determine the probability of a positive ion being reflected from a metal surface as a negative ion. His calculation was based on the following model. The ion forms an excited atom by extracting an electron from the metal. This excited atom then captures a second electron as it returns to
its ground state. The theory was applied to the reflection of mercury ions from nickel, for the energy states of the electrons in mercury ions and in solid nickel overlap conveniently.

Examining this theory, Sloane and Press (82) carried out a careful analysis of the particles emitted from a degassed surface, when bombarded with Hg\(^+\) and Hg\(^{++}\) ions. Detecting light negative ions but no Hg\(^-\) ions, they concluded that the light ions they observed were formed by the ejection of occluded material under the action of the ion bombardment.

Despite the fact that the existence of the Hg\(^-\) ion is in doubt, Smith's theory is still of some value. Assuming that the Hg\(^-\) ion did exist, he deduced its behaviour from its supposed electron affinity. Although the results are not meaningful in the case of Hg, the theory could well be applied to a case where the existence of the negative ion was better substantiated. However, for direct application to foil experiments the energy range of up to a few hundred electron volts is again too low.

Cobas and Lamb (21) calculated the probability of electron extraction from molybdenum, by He\(^+\) of a few hundred electron volts energy. Their results were later invalidated by those of Varnerin, who recalculated this extraction probability for energies up to 1000 eV, taking into account the image force (34, 66). The inclusion of the image force was found to decrease the calculated extraction probability.
In some more recent calculations (80), the neutralisation of ions and deactivation of metastable atoms has been treated in terms of field emission in the electric field of the ion or atom, the atom producing a field because of its dipole nature.

In 1963 an investigation of Auger ejection was made (75), in which the electron yield from polycrystalline tungsten was measured when bombarded with \( \text{He}^+ \), \( \text{H}_2^+ \), \( \text{N}_2^+ \) and \( \text{O}_2^+ \) ions at energies of 50, 100 and 200 eV. To obtain an atomically clean surface, the tungsten was heated at 1600°C for six days and then flashed to 2400°C in an ultra-high vacuum apparatus. The care that was taken to remove gas from the target indicates the importance of obtaining a very clean surface. For it was found that even minute quantities of gas on the surface influenced the electron yield greatly.

C. CHARGE EXCHANGE IN THIN FOILS

There have been very few charge exchange measurements made with foils and of these nearly all have been inconclusive because of the gas-covered targets used. With one exception, all investigations have been carried out on foils which had been exposed to the atmosphere in between evaporation and charge equilibrium measurement. The oxidation and gas adsorption that take place while the foil is exposed to the air are enough to obscure nearly all differences in equilibrium ratios that might otherwise be observed.

It is unfortunate that no work has been done on foils which were evaporated and bombarded in an ultra-high
vacuum environment. Not only would this ensure a very small amount of adsorption, but the composition of the foils would be much more consistent. For it has been established (38, 42) that the nature of foils, evaporated under ordinary high vacuum conditions, is affected by the composition of the background gases, and hence is very variable.

There has been no consideration of the effect of foil structure on the charge ratios. It has been tacitly assumed that the foils have laminar and amorphous structures, even though this is very seldom the case with evaporated films.

Only one calculation has been made of charge exchange specifically in metal. This was in 1924 when Fowler (32, 33) attempted to calculate the ratio of singly to doubly charged helium ions emergent from metal foils. Considering the electrons as a gas of temperature corresponding to the $\alpha$-particle velocity, he calculated the probability of capture by assuming that the $\alpha$-particles were in equilibrium with the gas and that a collision involving two electrons was necessary for electron capture. Electron loss he considered as ionisation by electron impact. As Oppenheimer (71) has pointed out, however, this theory has the disadvantage that the three body collision mechanism requires a capture probability varying as the square of the electron density, whereas experiment indicated a first power variation. Oppenheimer's contribution to the theoretical problem has been already outlined in Section A of this chapter.
The first experiments to resemble the present investigation, the measurement of equilibrium charge ratios emergent from foils, was made by Bartels in 1930 (9). As a part of his charge exchange experiments with hydrogen ions, he measured the charge ratios of ions emergent from celluloid foils (Figure 31). In these he observed that equilibrium between the positive and neutral components was always attained. In other words, his foils were always so thick (about 100 atoms) that variation of thickness made no difference to the ratio. This fact has been true for all charge exchange experiments in the energy range below 500 KeV, including the investigations in this thesis. Hence only a ratio of the capture to loss cross sections can be obtained and not the absolute values of the cross sections themselves. This is because, in a three component system, there are six charge exchange cross sections and measurement of the equilibrium ratios gives only two equations in these six unknowns.

In 1950 Hall (36) measured the ratio of charged to neutral components emergent from foils of beryllium, aluminium, silver and gold in the energy range from 20 to 400 KeV. He did not measure the negative component separately, but included it in the charged component. The foils he used were manufactured in a separate vacuum evaporation chamber and were transferred to the collision chamber, being exposed to the air on the way, with the deleterious effects already mentioned. Also, although liquid air traps were used, traces
of oil were sometimes found on the foil. In view of the sensitivity of the composition of the emergent beam to the nature of the last few atomic layers, it is not surprising that no significant difference between beryllium, aluminium and silver was detected. However, in the higher energy ranges, it was found that the gold foils produced a higher percentage of neutral particles. (Figure 29).

A year later, Ribe (76) made the first mention of the formation of negative hydrogen ions in solids. He was using silicon monoxide foils as windows for a gas cell, in experiments concerning the neutralisation of protons in hydrogen, and the negative ions were considered as a small perturbing influence on the measurements.

Alvarez (6) in the same year, suggested the idea of the tandem Van der Graaf accelerator, using foils for the production and the stripping of negative ions. He published a short table, giving the ratio of negative to positive deuterium ions emergent from foils of aluminium and silicon monoxide. The ratio was as high as 26% at 10 KeV and fell to 4.5% at 50 KeV. The foils were presumably contaminated, but in view of the practical aim of the experiment this was unimportant.

Some measurements with helium ions have been made by Dissanaike (25), who measured the ratios of He²⁺;He⁺;He equilibrated in beryllium, aluminium and mercury at energies between 130 and 1100 KeV. The foils were exposed to the atmosphere and no mention is made of liquid air traps. No differences in equilibrium charge ratios between the various foils were found.
Changes in the amount of the $H^+$ component as a function of time as (a) aluminum is evaporated onto the foil, and (b) gold and then aluminum are evaporated under identical conditions.

**Figure 6**
Equilibrium proton charge ratios from an aluminum foil contaminated with foreign material on its surfaces as a function of exit energy. (Several experimental points are included to show the probable error.)
H\(^+\) components as a function of the work function of the metals at several energies.
In 1953 Phillips (73, 74) reported the most satisfactory experiments yet carried out in this field. Realising the importance of surface contamination of the foils, he evaporated them in the collision chamber itself and surrounded them with liquid air traps. He observed a time rate of change in the ratios of the three hydrogen components while the foil remained in vacuo. Measurements were taken 16 seconds after the evaporation ceased. Except in the cases of the less reactive metals, gold and silver, the final ratio was obtained by extrapolating back to the time evaporation ceased, at which time the foil was considered to be clean. As an example, at 20.5 KeV, the beam emergent from aluminium contained 26% protons which rose to 38% in about ten minutes at $2.4 \times 10^{-5}$ torr (Figure 6). Phillips found the rate of this change to be proportional to the pressure. Definite differences in equilibrium charge ratios were detected between foils of different materials. However, the pressure in the collision chamber was still of the order of $10^{-6}$ torr, as measured by an ionisation gauge, and, according to calculations by Alpert (4), there must have been at least one monolayer of gas adsorbed on to the surface of the foil before the measurements started. One of Phillips' typical results is shown in Figure 7. A correlation which he obtained between the charge ratios and the work functions of the metals of the foils, is shown in Figure 8 and will be discussed in Chapter 5.

Phillips also did some experiments to see if the angle between the foil and the beam affected the results in any
way. His idea was to distinguish between bulk and surface effects of the type discussed in Section B of this chapter. No difference could be detected with foils which had been aged in the vacuum. In the case of fresh silicon monoxide and gold surfaces, small unpredictable changes of the ratios occurred, obscuring any systematic change with angle.

In the same year Fogel, Safronov and Krupnik (31) carried out experiments similar to those of Hall, measuring the ratio of positive to negative ions and ignoring the neutral component. They combined their results with Hall's to give a complete picture (Figure 35). Although their foils were exposed to the atmosphere before use, differences in charge ratios were nevertheless noted between different foils. In view of this, the combination of their results with those of Hall, which showed charge ratios to be almost independent of the substance of the foil, is hardly justifiable.

Recently, an experiment has been performed by Allison and Meckbach (60) to compare the effective charge of helium ions in solid and gaseous cadmium. To ensure that the measurements were independent of surface effects, they deduced the effective charge from the difference in stopping powers of the two phases of cadmium. The difficulties of making absolute stopping power measurements were avoided, by using the stopping powers for protons as a measure of the target thickness. The protons were assumed to have the same effective charge in the solid and the gas; hence the same stopping power
\[ Z_{He}^2 \gamma^2 = [\epsilon_{He} / \epsilon_H] \]
was taken to apply in both phases. This assumption was supported by the fact that the proton energies were always above 100 KeV, and hydrogen beams, equilibrated in hydrogen gas, contain 96% protons at this energy, while those emergent from an aluminium foil contain 98% protons. The stopping powers of solid and gaseous cadmium for both protons and helium ions were measured, the proton stopping power acting as a standard to within 2%.

The stopping of the He beam, which is a measure of the square of the effective charge, was found to differ by no more than 8% in the two phases (Figure 9). Thus the experiment indicates that the charge states of helium beams are not very dependent on the phase of the target material.

Use of the proton beam as a standard removes some point from the investigation, for it involves the assumption that the proton beam has the same charge composition in the foil as when it emerges. In other words, the absence of a large solid state effect was assumed in the case of hydrogen in order to prove that there was no large effect with helium. However, Allison intends to make absolute measurements of the helium stopping power and this will remove the above objection.

A theoretical comparison of the stopping powers of gaseous and liquid argon has been made by Neufield (65). The liquid argon was found to have a 12% higher stopping power because the increased density of this phase causes a polarizing field around the helium ions, the field being of sufficient magnitude to ionize completely any singly charged
ions that were formed in collisions. In this way the effective charge was increased in the condensed phase, which therefore had a greater stopping power. Allison and Meckbach do not compare this calculation with their experiment, because of the unsatisfactory estimates of the partial stopping cross sections for the individual charge states of helium, which would have to be used.

Another recent investigation of interest to the subject of charge exchange in foils, is the study of the effect of the crystal lattice on the stopping powers of solids (23, 64, 78). It has been ascertained that atoms with about 80 KeV energy can be channelled into low density planes in a crystal and can pass along these planes more readily than in a random direction. As the electronic environment of the fast particles is different in the low density planes, this could possibly affect the charge exchange processes as well as the stopping power. It would not be easy to measure charge ratios emergent from different crystalline faces in an experiment with thin foils, as the foil would have to be produced by epitaxial growth in a very clean vacuum system.
CHAPTER 2

PRINCIPLES OF EXPERIMENTAL PROCEDURE

INTRODUCTION

Described as briefly as possible, the present investigation consisted of firing protons through thin foils and analysing the undeflected emergent particles to find the ratio of $H^+ : H^0 : H^-$ over a range of energies.

In theory, the energy range over which the results of measurements would be interesting is between 0 and 150 KeV, this being the region in which the equilibrium ratios are most energy dependent. Over this range, the neutral component drops from above 50% to a few percent, while the negative component drops away from about 5% to less than 1%. But in practice, 5 KeV could be regarded as the lower energy limit, as below this the energy straggle induced by the foil would be proportionately very large. In this work the energy range lay between 20 and 120 KeV, the lower limit being set by the sensitivity of the detectors used, and the upper by the high tension set available.

The foils were made by evaporating a layer of metal on to a thin backing foil. The evaporated layer was on the side from which the particles emerged, the emergent face. This layer was of sufficient thickness (greater than 60Å) for equilibrium between the three charge states of hydrogen to be established and hence the backing foil had no effect on the charge composition of the emergent beam.

The choice of metal for foils was determined by
two factors; first, their suitability for forming evaporated layers and, secondly, their value for the comparison of charge ratios from different materials. Gold, silver, copper and aluminium were chosen, for these metals cover a wide range of atomic numbers, density and chemical reactivity and they all form evaporated films readily.

For the investigations the following components were required:

A. A proton beam of variable energy.
B. Target foils.
C. A means of analysing the emergent particles into the three charge states, and
   A means of measuring the emergent energies of the particles.
D. Detectors capable of measuring the relative intensities of the three charge states.

A basic consideration of these components will follow and the details of design will be given in the next chapter.

A. PROTON BEAM.

As every particle which suffered only a small angular deviation (\( \frac{1}{2^0} \)) on passing through the foil was counted in these experiments, as much as \( \frac{1}{3} \) of the beam incident on the foil finally reached the detectors. Therefore, if particle counting techniques were used, only a small beam current would be required. As explained in Section D, it was decided to use single particle detectors, one of the advantages being that the low beam current then necessary
enabled the ion source and beam focussing system to be of simple design.

1. **Ion Source.**

   A magnetic or Penning discharge ion source was chosen as simple to make and requiring no complicated power supplies (45). The fact that this ion source could be run from dry cells and accumulators was a very great advantage when the source was operated at high voltage, in that it avoided the use of large isolating transformers. The absence of R.F. fields was also advantageous when the detecting circuits were being set up, as the electromagnetic shielding was easier to arrange.

   The large energy spread of this type of ion source, often a drawback, was not serious in this case, as the spread was small compared with the energy straggle introduced by the foil.

2. **Incident Particle Selection.**

   The energy straggle also meant that no high resolution analysis of the incident beam was required. Hence a simple selector was used to remove the molecular ions and heavy ions such as $O^+$, $N^+$, from the beam. To avoid the necessity of altering the selector for different beam energies, it was placed close to the ion source; that is, the beam selection took place at low energy before the ions were given their main acceleration. This meant that a permanent magnet could be used to produce the fixed magnetic field for selection.

   Both a crossed electric and magnetic field
velocity selector and a $30^\circ$ magnetic momentum selector were tried successfully. The crossed field selector was used first as, when compared with the magnetic selector, it had the following advantages:

(a) The straight beam path was easier to align, and also the mechanical difficulties of an asymmetrical loading of the glass accelerating column were avoided.

(b) With the crossed field selector the total ion beam could be focussed and the analysis introduced later, whereas with the magnetic selector the two processes could not be separated.

(c) Although it was planned to use a permanent magnet for both selectors, the variable electric field in the crossed field selector permitted the ion type to be changed without trouble.

(d) The use of fairly strong crossed fields ensured that sufficient resolution could be obtained, whereas improvement of the resolution of the magnetic selector involved either a larger magnet or larger angle of bend, both of which were undesirable mechanically.

Preliminary experiments, in which the crossed field selector was used, revealed that the light from the ion source caused a considerable amount of noise in the electron
multiplier which was being used for particle detection. Hence, despite the several advantages of the crossed field selector, the problem of the ion source light made the change to the magnetic analyser desirable. For the bend in the beam path, while a disadvantage mechanically, proved of service, as it enabled the detectors to be shielded from the ion source light.

These preliminary experiments also showed that the beam intensity had to be reduced to achieve a counting rate low enough to be handled by electronic detectors. Thus the increased focussing difficulty which the magnetic selector entailed was no longer so important, as a highly focussed beam was no longer required. The change to the magnetic selector brought about a slight simplification of power supplies, with the elimination of the crossed electric field. The saving of voltage supplies was especially advantageous with the ion source at high potential, when all such supplies had to be isolated from earth.


All focussing was done by means of cylindrical electrostatic lenses (50, 84, 87). The first ion optical system was built up piece by piece and a well focussed ion beam produced. As already mentioned, the beam intensity obtained was found to be larger than necessary. Therefore, when the apparatus was later rearranged, the opportunity was taken to simplify the ion focussing system slightly by the omission of one of the cylindrical electrodes.
B. FOILS

When making the foils, two considerations were of prime importance. In the first place, owing to the low beam energy, it was essential for the foils to be thin. Secondly, the emergent surface of the foil had to be kept very clean, if the composition of the emergent beam were to be characteristic of the metal and not of absorbed gases and vapours.

1. Backing Foils.

Backing foils which were thin and yet robust and free from pinholes were required. They had also to withstand mild heating while located in the vacuum system during the outgassing of a zeolite cold trap. Plastic and alumina foils were tried and the alumina finally used, as the outgassing temperatures involved were sufficient to melt the plastic. The methods for making the backing foils are described in Appendix 1.

2. Evaporation of foils.

For the evaporated film to be representative of the bulk material, especially at the surface layers, the following conditions were necessary:

(a) The foil had to be evaporated in the target chamber so that there should be no contamination or oxidation in the atmosphere and so that the measurements could be taken immediately after evaporation.
(b) The evaporation should take place at about $10^{-9}$ torr to avoid large amounts of gas being absorbed into the foil structure during evaporation and adsorbed on to the surface afterwards.

The first condition was easily fulfilled, but not the second. Possibilities were investigated for keeping the foil in an ultra-high vacuum region in which the ion beam was to be generated (3, 5, 92); but as the time necessary to build such a system was thought to be prohibitive, an alternative plan was adopted. The vacuum system used in the experiment was quite conventional, being made of brass and steel sections put together with greased 0-rings, and pumped by an oil diffusion pump. The method used to achieve a better vacuum at the emergent side of the foil was to surround the face of the foil with a bed of artificial zeolite, which, after suitable outgassing, cooling caused to act as a pump (7, 37). This device (Figure 17) produced a high pumping speed, especially for oil vapour, in the vicinity of the emergent face of the foil. The pressure in the target chamber was reduced to $5 \times 10^{-7}$ torr, and it is likely that the pressure at the foil surface was of the order of $10^{-7}$ torr.

Although an improvement on the conventional high vacuum, this pressure is still rather high for the evaporation of reproducibly clean foils.

C. ANALYSIS AND ENERGY MEASUREMENT

The following two operations had to be carried out on the particles emergent from the foil:
(a) Analysis to enable the $H^+: H^0: H^-$ ratio to be measured.

(b) Measurement of the energy of the charged particles, normally the positive component only.

The emergent, rather than the incident, energy had to be measured because of the variable foil thickness. Measurement of the emergent energy of the protons also served to check the foils for pinholes, the presence of which would be revealed by a group of faster protons, which had passed through the foil without energy loss. Two methods of analysing the particles and of measuring their energy were considered, an electrical retardation method and a conventional magnetic spectrometer.

1. Electric Retardation.

It was hoped to be able to achieve the operations (a) and (b) given above by:

(i) measuring the emergent proton velocity by finding the retarding potential necessary to collect the protons at zero energy.

(ii) measuring the $H^+: H^0: H^-$ ratio by making each charge state strike the same detector with a different energy and separating the output pulses with a pulse height analyser.

It was proposed to have a layout resembling the Tandem electrostatic accelerator. The ion source and the detector were to be near earth potential and the foil between them at high negative potential. The protons were to be
accelerated from the ion source to the foil, then, after passing through it, decelerated as they travelled towards the detector. They could then be collected at zero velocity by making the detector a few KV negative to compensate for the energy lost in the foil. Since this negative voltage would be equal to the stopping power of the foil if the incident energy of the proton were known, through another electrical measurement, the emergent energy would also be known.

The reverse field between the foil and detectors would also achieve object (b). As a consequence of the field, the positive, neutral and negative particles emergent from the foil would strike the detector with zero, one and two times the incident energy respectively, the small amount of energy lost in the foil being subtracted in each case. So that the positive particles could be observed, the detector would have to be made about 25 KV negative, causing the protons to strike it with about 20 KeV energy; that is, with just sufficient energy to be registered.

It was intended to use an energy proportional detector with sufficient resolution to distinguish the three energies corresponding to the three charge states. A scintillation counter would have been a most suitable detector as the phosphor could have been set at -25 KV, the photomultiplier remaining always at earth, with a perspex light pipe providing insulation between the two. However, despite the advantages of this counter, its poor energy resolution
would probably have made it impracticable for use in this
application. If so, the scintillation counter would have
been replaced by a solid state counter in order to achieve
sufficient resolution to distinguish between the three sets
of particles. Since it is inconvenient to operate a solid
state counter at other than earth potential it would have
been necessary to give the protons an extra 25 KV acceleration
as they emerged from the ion source, by making the ion source
25 KV positive. A moveable Faraday cup would have been needed
for the energy measurement.


The alternative method for analysis and energy
measurement was to use a simple magnetic spectrometer. The
analysis was to be obtained by deviating the particles into
three separate detectors. An electromagnet was required,
which was capable of bending the positive and negative particles
in opposite directions through 90°, or some other suitable
angle. Sufficient space in the straight through position had
to be allowed for the detection of the undeviated neutral
particles. The resolution of the spectrometer did not need
to be very good because of the inhomogeneity of the beams
being analysed.


The two systems are compared below in a variety
of aspects.

(a) Mechanical Construction.

The electrical retardation scheme was simpler
mechanically, as no curved vacuum lines and heavy magnet were needed.

(b) **Electrical Circuits.**
In this regard the magnetic spectrometer had the advantage, in so far as no pulse height analysing circuitry was required.

(c) **Particle Collection.**
The electrical retardation method had the advantage of being able to collect particles from a large solid angle, as the detector was to be only about one inch from the foil. The collection angle of the magnetic spectrometer was much smaller, as the emergent particles had to be stopped down to a narrow beam before entering the spectrometer.

(d) **Target Chamber.**
There would be obvious disadvantages in having the foil in the high voltage terminal, as in the electrical retardation method. Any electrical circuits associated with the target chamber, such as the current supply for evaporation, would have to be disconnected before switching on the high tension.

(e) **Ion Source.**
Having the ion source near earth potential as in the electrical retardation method, would be an advantage from the point of view of providing its power supplies.
(f) Detectors.

The magnetic spectrometer allowed greater flexibility in the choice of detectors, as they need not have a response proportional to incident particle energy.

The simplicity of construction of the electrical retardation method was considered to be an important advantage, and the novelty of the scheme was also taken into account. Therefore it was decided to investigate the electrical retardation method further.

However, when the apparatus was set up, the method was found to have a fundamental objection, which had not been foreseen. It necessarily entailed having a high electric field near the detector, and the polarity of the field was such as to accelerate all stray electrons into the detector. Normally, similar high potentials between the electrodes of an accelerating gap would merely cause a very small electrical current and some X-ray production. But in the present case the detector formed one of the electrodes and the high potential caused a considerable number of large spurious pulses, even for the relatively low voltage of 20 KV.

This effect seemed to be due to the basic layout of the scheme and thus very difficult to rectify. Also the problem of obtaining a suitable detector had yet to be faced. Therefore, taking these two points into account, as well as the advantages of having an earthed target chamber, it was decided to make no further attempt to use the electrical
retardation method. The ion accelerating system was modified and the more conventional magnetic spectrometer was set up satisfactorily.

D. DETECTORS

In deciding what kind of detectors to use the following requirements had to be borne in mind:

(a) Ability to detect neutral atoms.
(b) Ability to detect low energy particles.
(c) A response independent of the charge state of the incident particle.
(d) Ability to detect very small beam currents.

The use of electrometers for making current measurements was precluded by (a). Current measurements, rather than particle counts, could also have been made with a phosphor and photomultiplier tube. However, to fulfil requirement (d), it seemed advantageous to use the system of phosphor and photomultiplier as a scintillation counter; that is, in the particle counting mode.

The use of particle counters would automatically ensure that the response of the detectors remains independent of the charge state of the incident particle, fulfilling (c). Also, small changes in detector efficiency would merely cause a change in pulse height, leaving the counting rate almost unaffected. This characteristic compares favourably with the current measuring mode of operation, in which changes in detector efficiency would be transmitted in direct proportion to the measurement of beam intensity.

Only with regard to (b), the low energy
requirement, did the particle counting method not seem the ideal answer. However, its advantages in all other respects were so great, that it was decided to accept a low energy limit of 20 KeV and use this technique. Not only did it fulfil three of the four requirements listed above, but it meant also that, as only a small ion beam was necessary, the beam heating of the target foil was reduced. A further advantage of a low intensity ion beam, already mentioned in Section A, was the simplification of the ion source and accelerator.

Electron multipliers, solid state counters and scintillation counters were available and the properties of each were investigated. The resolution between the signal and noise pulses from the electron multiplier was found to be very poor, but the other two detectors were satisfactory in this respect, for particle energies above 20 KeV. The small sensitive area of the solid state counter made it unsatisfactory except for neutral particles, as the charged particles spread too much in the magnetic spectrometer. Therefore a scintillation counter was used in the 90° position for the detection of the charged particles and a solid state detector in the 0° for counting the neutral particles. When the measurements were extended to low energies (20 KeV) this 0° solid state counter was replaced by a scintillation counter, in order to obtain slightly better low energy response. Another detector was considered briefly, the ion-electron converter (52), but was rejected because of its complexity.
A solid state counter was used at all times in the second of the 90° positions, where its small sensitive area gave the magnetic spectrometer sufficient resolution for the accurate measurement of energy.

Now that the basic principles of the apparatus and all major decisions have been discussed, we can proceed to the details of the particular parts of the equipment, which are given in the following chapter.
CHAPTER 3
DETAILS OF APPARATUS DESIGN

A. PENNING ION SOURCE

The ion source, which was based on a number of published designs (45, 49, 51), is shown in Figure 10, with the important parts numbered.

Hydrogen gas was admitted at (1), through a very slow leak consisting of a copper tube drawn tightly over a piece of stainless steel rod. The gas pressure in the source could be varied either by heating the copper tube, causing the space between the two metals to enlarge on account of the different rates of expansion, or by changing the pressure in the gas reservoir. This latter pressure was reduced to 20 cm.Hg for the normal mode of operation of the source, the copper being unheated. As knowledge of the exact operating conditions of the ion source was not important to the main investigation, the gas pressure in the source was never measured. However, it was known to be low by the small effect the ion source gas supply had on the vacuum conditions in the system. It was estimated that the hydrogen raised the system pressure by about \(5 \times 10^{-7}\) torr. The consumption rate of hydrogen was about 0.5 cm\(^3\) per hour (at 76 cm.Hg).

Three coils (2) produced an axial magnetic field in the discharge space (3). The aluminium anode (4), when at a sufficiently high voltage, depending on the magnetic field and gas pressure, maintained a column of plasma between
FIGURE II

FLUORESCENT SCREEN

FARADAY CUP

TO VACUUM PUMPS

ION SOURCE

PROBE
the two aluminium faced cathodes (5) and (6).

The cathodes were faced with aluminium to keep high the secondary electron production, which was necessary for the discharge, although the source could be operated with unfaced steel cathodes. With steel cathodes, however, a brown deposit built up on the glass, which quite quickly short circuited the anode to the cathodes. With the aluminium faced cathodes the deposit was grey and non-conducting. These deposits were obviously associated with sputtering of the cathodes. It seems that the aluminium was oxidized as soon as it sputtered, hence forming a dielectric layer of alumina. There was further evidence for sputtering of the aluminium face (5), in that it was eroded away in a central spot of about 1 mm. diameter.

Some of the positive ions from the discharge were accelerated towards the anode (6) and passed through the 0.020" aperture in it to form the ion beam. The ion current could be increased by adjusting the magnetic field near the exit aperture by means of small auxiliary magnets (45). The effect of these magnets was to centre the ion beam on the exit aperture, but, as sufficient ion current was already available, they were not used in normal operation.

For testing, the ion source was mounted on a 4" brass vacuum line, as shown in Figure 11. During this initial testing, higher gas pressures were used in the source and electron currents of 5 milliamps passed to the anode.
With these operating conditions, the ion current was easily measured with a spot galvanometer, and Figure 12 shows the typical variation of ion and electron currents with magnetic field.

When a small fluorescent screen was placed on the probe (7), which was earthed, the ion source could be seen to have a strong self-focussing action. A bright spot, about 1 mm. across, could be seen on the screen for various combinations of gas pressure and electric and magnetic fields.

The optimum operating conditions were as follows:

- Magnetic field: 200 gauss.
- Anode potential: 540 volts.
- Anode current: 100 m.a.

The anode potential was produced by six ninety-volt dry cells in series and a six volt lead accumulator provided about 2 amperes for the magnetic field coils.

B. FOCUSING, SELECTING AND ACCELERATING THE PROTON BEAM

The aim now was to produce a narrow beam of ions and to pass it through an analyser designed to transmit the protons only. Step by step construction was adopted, so that the progress of the ion beam could be followed throughout.

The first arrangement is shown in Figure 11. Various combinations of probe and cup potentials caused the ion beam to be focussed on to the fluorescent screen, which formed the end of the Faraday cup. The optimum focus was obtained with a probe potential of -250 volts and a Faraday cup voltage of -2000 volts.
FIGURE 13
The velocity analysing section was now added, as in Figure 13. It consisted of a 4" x 2" rectangular brass section containing a pair of parallel copper plates fixed 1 cm apart on glass and perspex insulators. A cylindrical electrode was included at either end of the plates. These two electrodes were to be held at a potential equal to the mean potential of the parallel plates, so that ions passing axially along the system would be travelling on an equipotential line. Hence the potential of these two electrodes determined the velocity of the ions passing through the selector. As the magnetic field in the crossed field analyser was fixed at 1200 gauss, by the permanent magnet available, the ion velocity had to be set between certain limits. Otherwise, an inconveniently large or small voltage across the parallel plates would be required to produce zero deflection on the protons. An energy of 3 KeV was chosen as suitable for the particles passing through the velocity analysing section, as this energy entailed an interplate potential of 730 volts.

Before the introduction of particle selection, the whole ion beam was focussed on to the screen. Therefore the magnet was not put in place and the parallel plates were short circuited together and held at -3000 volts with electrode 2 and the Faraday cup. With this arrangement it was possible to focus the ions on to the fluorescent screen by raising the potential of electrode 1 to -2500 volts.

Velocity analysis could now be introduced. A permanent magnet, with a 1200 gauss field in the 2" gap between
its 2" square plates, was placed so that its magnetic field was perpendicular to the electric field between the parallel plates. A variable D.C. supply provided the voltage between the plates and this supply was so arranged that the mean potential of the plates was always -3000 volts. The interplate voltage was varied to allow the different ion species to be focussed on to the fluorescent screen in turn. The magnetic field in the analyser was constant and the ions entering had all been accelerated through the same potential (-3000 volts). Therefore the square of the interplate potential was proportional to the charge-to-mass ratio of the ion species passing undeviated through the analyser.

The table below shows the results obtained with the velocity analyser, V being the interplate voltage and the ion currents being in arbitrary units.

<table>
<thead>
<tr>
<th>V</th>
<th>730</th>
<th>514</th>
<th>410</th>
<th>165</th>
<th>120</th>
</tr>
</thead>
<tbody>
<tr>
<td>\left(\frac{730}{V}\right)^2</td>
<td>1</td>
<td>2.02</td>
<td>3.17</td>
<td>19.6</td>
<td>37.0</td>
</tr>
<tr>
<td>Ion type</td>
<td>H⁺</td>
<td>H₂⁺</td>
<td>H₃⁺</td>
<td>O⁺, N⁺</td>
<td>O₂⁺, N₂⁺</td>
</tr>
<tr>
<td>Current</td>
<td>4</td>
<td>50</td>
<td>12</td>
<td>3</td>
<td>2</td>
</tr>
</tbody>
</table>

As protons were the particles required, the potential between the parallel plates was fixed at 730 volts. Since it was desirable to re-focus these protons before acceleration, an extra electrode, whose voltage could be altered arbitrarily, was included in the next stage. At the same time the 12" long glass accelerating column holding the high voltage electrode was added (Figure 14). As the fluorescent screen was now at high potential, the focal spot
FIGURE 15
was viewed from a distance by means of a mirror. The protons could be focussed on to the screen for high tension voltages between 10 and 120 KeV by varying the voltage on the probe and the electrodes 1 and 4.

A well focussed proton beam was now available for use. Nevertheless, when the system was inverted, and the ion source put at the high potential end, the opportunity was taken to introduce two modifications to the focussing arrangements. It will be recalled from the previous chapter that the inversion of the system was brought about by the change from the electrical retardation method of particle analysis to the magnetic spectrometer.

Firstly, the crossed field velocity selector was replaced by a magnetic analyser, so that light from the ion source might be kept from the detectors, as explained in Chapter 2. The other change was the omission of one of the variable potential electrodes and its power supply, as it was now realised that sufficient proton intensity could be achieved with a more simple focussing system. The modified system is shown in Figure 15.

Thirty degrees was chosen as the angle through which the proton path bent in the magnetic analyser because:

(a) A $30^\circ$ bend would give sufficient resolution for the elimination of the unwanted ions from the beam.

(b) A larger bend would introduce mechanical difficulties, in the form of a large torque
Figure 16

SHIELD

MAGNET COIL

640 V.

PROBE

MAGNETIC FIELD

10 KV D.C. CONVERTER

0 to 150 KV POWER SUPPLY

TARGET CHAMBER

ANALYSER AND DETECTORS
on the glass column supporting the ion source.

(c) It was convenient to use the same 1200 gauss permanent magnet. The ion energy for a 30° bend between the poles of this magnet was suitable, being about 7 KeV.

The optimum proton energy was later measured to be 8.5 KeV. In order to give the protons this energy, the ion source was insulated from the analysing section by a perspex ring, and made 8.5 KV positive.

The beam diameter was controlled by apertures and the intensity adjusted to suitable levels by varying the voltages on the probe and electrode 1. Electrodes 2 and 3 were solely to guide the protons into and out of the equipotential magnet box, so that no electric forces were experienced in this region. The beam intensity was measured with an electron multiplier and the magnetic spectrometer (Section D) was used to ascertain that the proton intensity was maximised and the unwanted ions eliminated. A typical beam profile is shown in Figure 23.

The layout of the power supplies for the ion source and for the focussing and accelerating voltages is shown in Figure 16. The ion source, being at the high tension end of the accelerator, was powered by dry cells and an accumulator, as described in Section A.

Similarly, a battery driven 10 KV D.C. converter supplied the voltages for the focussing electrodes and the 8.5 KV potential on the ion source. The D.C. converter was
powered by a 12 volt lead accumulator. Appendix 2 gives the details of its circuit. The 10 KV output of the converter produced a current of 100 μa. in a multi-resistor voltage divider. The 8.5 KV for the ion source potential was tapped from the chain in a soldering iron adjustment. The variable voltages for the probe and electrode 1 were selected from the chain by means of multiposition rotary contacts. To enable these switches to be operated while the high tension was on, they were turned by means of long tubes of bakelized cardboard.

All the batteries, the D.C. converter and its resistor chain and switches, the ion source gas reservoir and the permanent magnet for the beam selector were mounted on a wooden table supported by bakelized cardboard legs. The evacuated accelerating column passed through a hole in the table.

The 0-120 KV accelerating voltage was provided by a Kilovolt corporation 150 KV set. It was a conventional 50 cycle per second transforming, voltage multiplying and rectifying system. The ripple at no load, which was its mode of operation in its capacity as the acceleration voltage supply, was measured and found to be ½%. Although the set was designed with an output of 150 KV, electrical breakdown in the oil insulated transformer prevented operation above 120 KV.

Two precautionary measures were necessary to prevent the high tension from damaging the D.C. converter,
in the event of corona, brush or vacuum discharges. Firstly, the high tension was connected directly to electrode 3 (Figure 15), so that the current for any discharge in the accelerating gap would come directly from the high tension set, and not through the D.C. converter.

Secondly, all equipment on top of the insulated table was enclosed by an aluminium shield, which was connected directly to the high tension voltage. Again, if any discharge to earth took place, it was directly from the high tension set. The set itself was unlikely to be damaged, owing to its large current rating of 2.5 m.a. and its effective overload protection.

The aluminium shield was made of flat panels, which could be removed easily for routine maintenance, such as battery charging and the filling of the gas reservoir with hydrogen. The shield was also made smooth to reduce corona current which would have been a great source of electric noise, making the shielding of the detecting circuits more difficult.

A useful proton beam was now available at earth potential in the target chamber.

C. TARGET CHAMBER

The collisions between the protons and the atoms of the foil took place in a steel target chamber, 8" diameter, 12" long, which was designed to contain:

1. A number of backing foils held so that each could be moved into the beam path in turn,
FIGURE 17

PROTON SEAM
LIQUID AIR INLET
MIRROR PROBE
ZEOLITE COLD TRAP
APERTURE OF MAGNETIC SPECTROMETER
ION GAUGE
HEATED FOIL CUP
ELECTRON EJECTION FILAMENT
VACUUM VALVE
FIGURE 17
without admitting air to the vacuum.

2. The zeolite cold trap.

3. A small thoriated tungsten filament for bombarding the backing foils with electrons before evaporation.

4. Evaporation filaments loaded with metal for coating the foils.

The following standard items were also associated with the target chamber:

a) A Veeco Bayard-Alpert ionization gauge.

b) A vacuum valve for fast pumping during degassing of the zeolite.

c) A lamp and observation windows.

1. Foils

The details of manufacture of the backing foils are given in Appendix 1. Each foil was mounted on the bottom of a 1" long, 1" diameter brass cup by means of a stainless steel plate. Six of these cups were mounted at a time in holes in a brass wheel in the target chamber (Figure 17). One end of the steel axle of the wheel passed through the wall of the target chamber, and was sealed with an O-ring. Thus the wheel could be rotated, bringing each foil into the beam path in turn. The wheel could also be raised and lowered, by sliding the axle up and down. This freedom of movement enabled the foil cups to be placed in the well of the zeolite cold trap. In this way the foil was located in the beam path and at the same time was surrounded by absorption pump material.
Gold Trap, Evaporation Probe and Filaments

Figure 18
For some experiments it was necessary to heat the foils. To achieve this, the backing foil was mounted in a stainless steel cup and held in place by a small steel disc with a heating tape wound in it. The performance of the heater was tested in a separate vacuum system, in which it was found that the foil could be heated to about 300°C, with a heater current of 1½ amperes. At temperatures above 300°C the strains set up by the heat very often broke the alumina backing foil.

2. Zeolite Cold Trap

The special cold trap surrounding the foil consisted of a bed of artificial zeolite pellets, contained between an outer steel cylinder and an inner piece, made of molybdenum wire in an open mesh structure (Figures 17 and 18). The inner wire mesh cylinder formed the well into which the foil cups fitted. A central mesh tube of smaller diameter (¼") led from the bottom of the well and formed the beam path for the particles emerging from the foils. This smaller mesh cylinder also provided the evaporating metal with access to the underside of the foil.

By this means, the emergent surface of the foil was surrounded by zeolite, which absorbed gases through the mesh.

Embedded in the zeolite was a coil of tungsten wire of 1 mm. diameter, insulated with glass braid. During the outgassing of the cold trap a current of 10 amperes through the tungsten coil heated the zeolite to 300°C, as
measured by a thermocouple. The temperature could not be increased much beyond 300° because of the excessive heat that would be conducted to the neoprene O-rings sealing the target chamber. After 12 hours outgassing the vacuum in the target chamber was about $5 \times 10^{-6}$ torr when the trap was still hot, and $1 \times 10^{-6}$ torr after it had cooled to room temperature.

During the evaporation of the foil and the measurement of its charge ratios, liquid air was circulated in a coil of copper tubing that surrounded the zeolite. Thermocouple measurements indicated that the zeolite was cooled nearly to liquid air temperature. The pressure in the target chamber fell to $5 \times 10^{-7}$ torr. Both ends of the copper coil passed through the wall of the target chamber inside thin-walled, stainless steel tubes, to reduce the heat conduction from the chamber. The liquid air was forced slowly through the coil by air pressure.

Although it would have been useful to know the pressure close to the foil surface, no vacuum gauge was put in this position, because of the desirability of making the cold trap enclose the emergent face of the foil as completely as possible.

3. **Electron Bombardment**

When a foil was in the well of the cold trap, one of the adjacent foils rested just above a small thoriated tungsten filament. Each backing foil was put in this latter position and bombarded with a few milliamperes of electrons just before being placed in the cold trap for evaporation and
measurement. The electron bombardment degassed the foil, hence preventing formation of pinholes in the metal layer subsequently evaporated on to it.

The effectiveness of this procedure was seen when foils, made both with and without electron bombardment, were compared under a transmission microscope. The films deposited on outgassed foils had very few holes, about two or three in a foil of 3 mm. diameter. On the other hand, foils deposited without prior degassing had a few hundred pinholes in the same area.

4. Evaporation of Foils

A photograph of the evaporation probe is shown in Figure 18. It can be seen that the probe carried three filaments; each was used separately, being rotated in turn to the uppermost position. All the filaments were charged with the same metal, three being used to ensure that there was sufficient metal to coat six foils.

The whole probe could be slid back and forth in the target chamber. For evaporation it was positioned so that the blob of molten metal was directly beneath the beam tube running through the zeolite cold trap to the foil. It was found that no air was admitted to the vacuum as the probe slid outwards, but that as it returned, the pressure rose momentarily. This pressure rise was due to the entry of a gas-covered, greased surface into the system. The probe was withdrawn from the vacuum after evaporation of a foil; that is, just before the charge ratios were measured. It was
important that the probe never moved inwards at this stage, otherwise air would be admitted to the system, contaminating the fresh surface of the foil. Hence, a counterweight was used to balance the atmospheric pressure, thereby ensuring that the probe was in equilibrium in all positions.

A small metal mirror and viewing port were mounted on a similar probe above the foil. With the aid of this probe, the molten metal could be positioned by sighting it through the foil. The viewing probe also enabled an estimate of film thickness to be made, by permitting observation of the incandescent filament through the backing foil. The filament appeared to darken as the metal condensed on the foil.

Two types of filament were used. For aluminium, the filaments were made of a piece of 1 mm. diameter tungsten wire, bent as shown in Figure 18. The bends were put in to increase the length of wire between the hot metal and the filament supports, thereby minimising heat conduction losses. The other three metals, silver, gold and copper, were evaporated from small conical helical baskets of triple stranded 0.020" molybdenum wire (Figure 18). These baskets were used to counteract the tendency of the metal to drip off the filaments. A steel cradle hung below the filaments to catch any such drips and to prevent evaporated metal from blocking the aperture to the magnetic spectrometer.

The filaments were heated thoroughly before the evaporation of a foil, in order to outgas all parts of the
system exposed to the heat. This heating was carried out with the probe fully inserted, so that the hot metal was out of the line of the foil. If the outgassing was thorough, the metal could be evaporated on to the backing foil at a pressure below $10^{-6}$ torr. Currents of up to 30 amperes A.C. were used to heat the evaporation filaments.

The molten metal could be seen through an observation window in the chamber wall. The filament was not in direct line with the window but was viewed indirectly in a mirror, to avoid coating the window with evaporated metal.

The following procedure was adopted before measurement of the charge ratios on a fresh foil:-

a) A cold finger trap above the diffusion pump was filled with liquid air, bringing the pressure down to $10^{-6}$ torr.

b) An alumina backing foil was bombarded with a few milliamperes of electrons at 100 volts.

c) Liquid air was circulated through the coil of the zeolite cold trap, bringing the pressure down to $5 \times 10^{-7}$ torr.

d) The backing foil was moved into the well of the zeolite cold trap.

e) The evaporation filament was heated in the fully inserted position, sufficient time being allowed for the pressure to stabilize, usually at about $8 \times 10^{-7}$ torr.
f) The evaporation probe was withdrawn until the molten metal could be seen through the foil, by means of the mirror probe.

g) When slight darkening of the foil indicated that some metal had deposited on it:-

i) A stop-clock, used to measure the age of the foil, was started.

ii) The evaporation current was turned off.

iii) The evaporation and mirror probes were withdrawn.

iv) The windows in the target chamber were covered to keep light from the detectors.

v) The high tension was turned on and measurements of the charge ratios started.

D. MAGNETIC SPECTROMETER

The magnetic spectrometer used for analysis of the emergent particles was small and simple (Figure 19). The pole faces were 10" x 5" and the gap width 1". The radius of curvature of the charged particles was 10 cm. and the entrance aperture 0.5 mm. diameter. The particles entered the spectrometer centrally, so that the positive and negative ions could be deviated through 90° in opposite directions. The neutral particles could pass straight on through a 2" square hole in the magnet yoke. Because of this neutral beam passage, the coils of the magnet had to be wound round the pole pieces, rather than the yoke.
A 50 volt, 10 amp variable D.C. supply was used to energise the magnet. As the supply was voltage stabilized, corrections to counteract current drift, caused by temperature changes in the magnet coil, were made manually.

The magnetic field was measured with a Hall probe gaussmeter. By this means all difficulties due to hysteresis were avoided. The Hall probe was located in the magnetic field by a small slot fixed between the pole faces. The slot enabled the probe to be returned to exactly the same position after removal for recalibration.

E. ENERGY CALIBRATION OF SPECTROMETER

The magnetic spectrometer was calibrated for energy measurement by accelerating a beam of protons through a series of known voltages and correlating these voltages with the magnetic fields necessary to deflect the protons through $90^\circ$. No foil was in the beam path and hence there was no energy loss in the beam. A solid state counter with a small sensitive area ($\frac{1}{2} \text{ cm}^2$) was used in one of the $90^\circ$ positions, to detect the peak counting rate of the deflected protons. The amplified output of this detector was measured on a vacuum tube voltmeter.

The absolute accuracy of the Hall probe gaussmeter was of no importance to the accuracy of the energy calibration. In fact, owing to the inhomogeneity of the magnetic field, the gaussmeter reading depended on the arbitrary position of the probe. The important point was that the position of the probe and the calibration of the
meter remain constant once the spectrometer had been calibrated.

To find the potential through which the protons were accelerated, the high tension voltage was measured. To do this, a voltage divider, consisting of thirty 100 MΩ resistors connected in series with a 12 MΩ resistor, was connected between the high tension and earth. The total resistance of the chain was R (nominally $3 \times 10^9 \Omega$). The test resistance r (nominally 12 MΩ) was at the earthed end of the chain.

When the high tension was set at any particular value, V, the voltages V, v corresponded to the resistance R, r such that

$$\frac{V}{v} = \frac{R}{r}$$

or

$$V = v \frac{R}{r}$$

To measure the high tension voltage V, the voltage v was measured by means of a Fluke differential voltmeter. This device worked by balancing the external voltage against an internal reference potential and therefore, when balanced, left the external circuit unaffected. Readings of v and of the corresponding magnetic fields for 90° proton deflection were taken for energies between 0 and 120 KeV. This energy range corresponded to values of v from 0 to 500 volts, which was the full range of the differential voltmeter.

It was then necessary to calibrate the voltage divider, or, in other words, to measure $\frac{R}{r}$. This was done by
disconnecting the high tension set and using a Fluke 500 volt supply to produce the voltage V. The differential voltmeter was now used to measure both V and v for a few values of V and the table below shows some typical results.

<table>
<thead>
<tr>
<th>V volts</th>
<th>450.77</th>
<th>491.90</th>
<th>450.62</th>
</tr>
</thead>
<tbody>
<tr>
<td>v volts</td>
<td>1.84</td>
<td>2.00</td>
<td>1.836</td>
</tr>
<tr>
<td>( \frac{V}{v} = \frac{R}{r} )</td>
<td>245.0</td>
<td>245.9</td>
<td>245.4</td>
</tr>
</tbody>
</table>

As a check, the voltage V was alternatively obtained from an accurately calibrated Fluke 6 KV supply. Results from these measurements agreed to within \( \frac{1}{2} \% \) with those above. The results of the first method were used, as the accuracy of the measurement of \( \frac{R}{r} \) then depended only on the linearity of the differential voltmeter and not on the calibration of the voltmeter or any power supplies.

In order to obtain the proton energy, the value of the focussing voltage provided by the D.C. converter had to be added to V, the high tension voltage. This potential was measured to be 8.5 KV with a Sweeney electrostatic voltmeter, which had been calibrated against the Fluke 6 KV supply. The stability of the D.C. converter voltage affected the accuracy of energy calibration only during the calibration measurements. Once these were complete, fluctuation in the 8.5 KV had no effect on the energy measurement, for the energy was measured by means of the magnetic field only.

A source of error arose from the fact that the protons were ejected from the ion source with a finite velocity. The maximum ejection energy was 540 eV, given by the potential
Pulse Height Distribution for Electron Multiplier with 30 KeV Protons

Figure 20
of the anode, but in fact the protons probably emerged from the ion source with about 200 eV, because of the potential drop in the discharge. 0.2 KeV was added to the proton energy and an error of 100% allowed in this addition.

An internal check of the measurements was provided by the fact that the graph of $B^2$ versus the proton energy was a straight line passing through the origin. The energies of the particles in later experiments were calculated from

\[ E \text{ (KeV)} = kB^2 \quad (B \text{ in Kilogauss}) \]

where \( k = 4.59 \text{ KeV/(KG)}^2 \)

F. DETECTORS

1. Electron Multiplier

The electron multiplier used was an E.M.I., 9603, 13 stage, venetian blind particle counter. This tube was found to respond to the lowest energy ions available which were at 8.7 KeV. A typical pulse height distribution curve of one of the 9603 multipliers is shown in Figure 20. The figure also shows a curve obtained from a tube which had aged for some weeks in the vacuum. These curves were taken with a high gain, low noise amplifying system, in an effort to resolve the signal and noise pulses. As can be seen from the figure, this effort was not successful. In fact, very large differences in counting rate could be obtained by small shifts in the lower threshold of pulse height.

This counter was used for early work in setting up the ion beam, and in plotting beam profiles with the magnetic
spectrometer. For all later measurements it was replaced by scintillation counters and solid state detectors because of the poor pulse height distribution described above.

2. Solid State Detectors

ORTEC 20A30 silicon, surface barrier detectors were mounted in the $0^\circ$ and $90^\circ$ positions of the spectrometer. The output pulses were amplified by ORTEC 201-101 amplifiers and pre-amplifiers. The pulse height distributions for various proton energies and bias voltages are shown in Figure 21. It can be seen that the signal and noise pulses can be resolved for energies above 25 KeV, which is in good agreement with a previously reported measurement of resolution (26).

When the counting rates for the $0^\circ$ and $90^\circ$ detectors were compared, it was found that the $0^\circ$ counter consistently had a higher rate. This fact was not unexpected because of the small surface area of the counters. The $0^\circ$ count was taken with zero magnetic field, the beam spreading, therefore, being purely geometric. In the $90^\circ$ case, however, further beam spreading was caused by defocussing in the magnetic field. The effect was more marked if the beam had passed through a foil, for the energy straggle it induced caused yet further spreading of the beam. Thus, because of the small detector size, many particles were lost in the $90^\circ$ position, which meant that a solid state counter could not be used for detecting the total charged component.

3. Scintillation Counter

Because of the disadvantage of the small area
PULSE HEIGHT ANALYSIS - SCINTILLATION COUNTER

Figure 22
of the solid state counters, a scintillation counter was set up in the other 90° position, opposite the 20A30 counter. The phosphor was thallium activated Cs I, 1" square and bonded to a perspex light pipe with epoxy resin. The light pipe formed the vacuum seal, and an E.M.I. 9614S photomultiplier was coupled to the perspex with paraffin oil.

The output pulses were amplified by a Dynatron Radio cathode follower and amplifier. Figure 22 shows the pulse height distributions of the output for various incident proton energies and multiplier voltages. The multiplier potential normally used was 1400 volts. At this voltage, as can be seen from Figure 22, the pulses from 20 KeV protons could be distinguished from the noise. Slightly better resolution than this has since been reported (77).

The proton beam was swung back and forth, by means of the magnetic field, between the 0° 20A30 and the 90° scintillation counter. The counting rate of the two counters agreed to a satisfactory extent (± 2%), even with a foil in the beam path (see Section A4 Chapter 4). Thus it was shown that the scintillation counter had a sufficiently large surface area to record all the particles in the 90° position.

The arrangement, consisting of two 20A30 counters and the scintillation counter, was used for many measurements. The 20A30 remaining in one of the 90° positions was used as the detector when the magnetic spectrometer was used for energy measurement. For this application the small surface area was advantageous, in that the resolution of the spectrometer was
increased.

At a later date the 0° solid state detector was replaced by another scintillation counter in order to achieve slightly better low energy response. This counter was very similar to the first scintillation counter except that a 1" diameter photomultiplier (E.M.I. 9524S) was used because of the lack of space. This smaller tube had a box and grid structure in place of the venetion blind dynodes of the 2" tube. However, the pulse height distributions of the two tubes were found to be very similar.

We have now covered all aspects of the design of the apparatus and, after a discussion of the factors limiting the accuracy of the measurements, in the next chapter, we can proceed to the experimental results in Chapter 5.
CHAPTER 4

EXPERIMENTAL ACCURACY

INTRODUCTION

Fluctuations in results due to the limitations of the apparatus only are considered here; any variation of the measured charge ratios caused by differences in the foils will be treated in the next chapter as a part of the results.

The factors contributing to the experimental error are discussed in Sections A, B and C. These are followed by a consideration of the reproducibility of the measurements and finally by an estimate of the overall accuracy of the experiment.

A. GENERAL

1. Composition of Proton Beam

Magnetic analysis of the primary beam revealed a small number of $\text{H}_2^+$ ions as the only impurity. In passing through the foil, these particles dissociated to form two atomic hydrogen particles, each with one half the energy of the particles that were accelerated as protons. These slow particles then attained the charge equilibrium composition characteristic of their energy and the material of the foil. The charged component formed from the dissociated molecules was not counted with the more energetic charged particles, as the magnetic field did not focus them into the 90° detectors. However, because of the absence of any energy selection for the neutral component, the fast and slow atoms were counted together.
Figure 23

BEAM PROFILE

Particle Counts per 30 seconds.

H⁺ (Primary)

Half width 23 KeV

H⁺ x 10
(From foil)

Half width 34 KeV

H₂⁺ x 100 (Primary)
The molecular ions would have constituted a serious source of error if the number present had not been shown to be very small (less than 0.1%) compared with the number of protons. This was demonstrated by removing the foil from the beam and analysing the primary beam with the magnetic spectrometer. A typical beam analysis curve is shown in Figure 23.

When measurements were made on the primary beam, its intensity was reduced by 400 times, so that the counting rate was not excessive. This reduction of intensity was achieved by defocussing the beam. The proton intensity was reduced by a larger factor than the $\text{H}_2^+$ ion intensity by this procedure, as the molecular ions were scattered, rather than focussed, into the beam. Therefore, as the intensity of the molecular ions was less dependent on the focussing conditions than that of the protons, the proportion of molecular ions present in the full intensity beam, used for foil bombardment, would have been less than the 0.1% measured by the method described.

A further check was afforded by the magnetic analysis of the charged particles emergent from a foil. If any low energy charge particles were present, their intensity was so small as to be indistinguishable from the background count.

2. Charge Exchange in the Residual Gas

There are three regions in which the beam could undergo charge changing collisions with gas molecules remaining
in the vacuum.

1) Between the ion source and the main accelerating gap. Any of the slowly moving ions neutralised in this region would not be accelerated. Most of them were lost from the beam in any case, as the electric and magnetic fields for focussing and selecting did not operate on them.

2) Between the accelerating gap and the foil. Any atoms or negative ions formed in this region would leave the results unaffected, as the equilibrium ratio formed in the foil was independent of the charge state of the incoming particles.

3) Between the foil and the detector. This was the only region where gas collisions had to be considered carefully, for if one of the charge components underwent an excessive number of charge-changing collisions on the way to the detector the counting rates could be seriously modified.

However, it can be easily shown that this effect is negligible. The largest cross section, which could have affected the results of the investigations, is that for loss of the second electron from the negative ions, and is denoted by $\sigma_{-2}$, where

$$\sigma_{-2} = 10^{-15} \text{ cm}^2 \text{ per atom} \ (1).$$
The proportion of particles undergoing a charge-changing collision with a gas molecule is given by \( R \) where

\[
R = \frac{N}{760} \sigma T_o l p
\]

where \( l = \) path length between foil and detector
\( = 20 \) cm.

\( p = \) background gas pressure
\( = 10^{-6} \) torr

and \( N = \) no. of molecules \( \text{cm}^{-3} \) at 760 torr
\( = 2.6 \times 10^{19} \)

therefore

\[
R = 6 \times 10^{-4}
\]

That is, one in every 1600 particles underwent a charge-changing collision in the gas. Hence the residual gas had negligible result in this regard.

Phillips, in his experiments, found that changing the pressure from \( 5 \times 10^{-5} \) to \( 10^{-4} \) torr, changed the charge ratio by no more than \( 1\% \), which is in agreement with this calculation.

3. Charge exchange and scattering at the aperture

It was possible that the 0.5 mm diameter aperture between the foil and the magnetic analyser could have influenced the results if:-

a) The aperture caused excessive scattering of the beam, or

b) A large number of charge-changing collisions could take place at its surface.
The aperture, which was made of mild steel, was tapered to a knife edge to minimise these effects. A test for scattering was made by analysing the beam with the magnetic spectrometer with no foil in position. The low background count away from the peak indicated that only a small amount of scattering (less than 0.1%) occurred. The beam profile is shown in Figure 23.

A further test showed how few charge exchange collisions took place at the aperture. This test consisted of a measurement of the neutral counting rate, with no foil in position, using the low intensity, defocussed beam. The neutral count was found to be 0.25% of the total beam. Neutral particles formed by gas collisions (as just discussed in Part 2) were also included in this count, showing that the estimate of neutral particles produced in the gas is justified.

4. Losses in the Magnetic Spectrometer

Once the particles had passed through the 0.5 mm aperture at the entrance of the magnetic spectrometer, it was important that all of each component should reach the appropriate detector. Some of the particles may have struck the walls of the vacuum chamber before reaching the detectors, due to:

a) Defocussing of the charged particle beams in the magnetic field.

b) Divergence of the charged particles caused by their energy spread.

c) Geometrical divergence of either charged or
neutral components.

d) Asymmetry of the apparatus, causing the beam to deviate from the central axis of the system.

e) Incorrect magnetic field for the particle energy involved.

A test to determine the importance of the total of all these effects was made by taking beam measurements with a foil in place. The foil was introduced for this test because the energy straggle it introduced increased the divergence due to energy inhomogeneity (b), and hence increased the demands made on the apparatus. By alternately setting the magnetic field to zero and to the value B gauss, corresponding to the energy of the beam, the positive component was measured in the 0° counter and the 90° counter in turn. Thus with zero magnetic field, the total beam was measured in the 0° detector. With the field B, the total beam was found by summing the counts from both detectors and making a correction of a few percent, dependent on the energy, for the loss of the negative particles, which were not counted. The measurement was carried out for the range of energies and was repeated several times in each case to allow for fluctuations in the primary beam intensity. The test was repeated from time to time throughout the course of the measurements to check the operation of the apparatus. Except when an excessively thick foil was used, with stopping power greater than 10 KeV, the two total count measurements never differed by more than 5%. Although this was the greatest source of
doubt in the measurements, it was considered to be satisfactory.

B. DETECTORS

1. Noise

The spurious count in the scintillation counters, primarily caused by photomultiplier noise, was easily recorded when the ion beam was turned off or stopped by a solid target. The noise count remained small (3 counts per second) and constant, enabling the particle counting rate to be easily corrected when necessary. In general, the counting time was 30 seconds, during which time more than 20,000 counts were collected in each detector. The spurious count over the same period of time was about 100. Hence, even without correction, the noise contributed less than 0.5% to the error. The noise from the solid state counter was 100 times smaller than that from the scintillation counters and thus was negligible at all times.

In the case of negative ions, with much lower counting rates, the noise correction of 3 counts per second in the scintillation counter was more important and had to be subtracted from the negative ion count.

2. Counter losses due to high counting rate

For the measurements with positive ions, the counting rate did not exceed 60,000 counts in 30 seconds. With a pulse length of 2 μs and a scaler paralysis time of 1 μs, the maximum correction for counting losses was given by

\[
k = \frac{\text{Counting time}}{\text{counting time} - \text{Total count} \times \text{dead time}}
\]
Hence it can be seen that the error due to counting losses is less than 1%.

The counting loss was a more serious consideration during the comparison of neutral and negative counting rates. The difficulty here was caused by the wide difference in counting rate of these two components - differences of up to 100 times. To collect $10^4$ negative ions, up to $10^6$ hydrogen atoms had to be counted. The counting time was extended to 90 seconds for these measurements and an accurately calibrated 5 µs paralysis time was introduced in the scaling circuit.

The correction was now

$$\frac{90}{90 - 10^6 \times 5 \times 10^{-6}} = 1.06$$

Hence in the investigations with negative ions, a counting loss correction of up to 6% was applied to the measurements of the neutral particle counting rate.

3. Effect of Stray Magnetic Field on Detectors

It was essential to verify that the magnetic field did not adversely affect the counting efficiencies of the photomultiplier tubes. Two approaches were adopted:

(a) Measurement of the magnetic field in the tube positions, and

(b) Measurement of counting rate with different fields.

(a) A Hall probe gaussmeter was used to measure the
EFFECT OF MAGNETIC FIELD ON PHOTOMULTIPLIER

VENETETIAN BLIND STRUCTURE—PHOTOMULTIPLIER TYPE 6097B.

1. FIELD FROM ANODE TO CATHODE
2. FIELD FROM CATHODE TO ANODE

BOX & GRID STRUCTURE—PHOTOMULTIPLIER TYPE 9524 B

Figure 24
field inside the mild steel magnetic shields of the two photomultiplier tubes. The fields were found to be approximately 2 and 6 gauss, for the 90° and 0° detectors respectively. These stray fields were both measured with a 5000 gauss field between the poles of the magnet, the maximum field used.

Data given by the makers of the photomultiplier tubes, E.M.I., indicated that the 2 gauss field would have very little effect on the efficiency of the venetian blind tube used in the 90° position. (Figure 24). On the other hand the data indicated that, when a box and grid tube was used in the 0° position, its orientation in the 6 gauss transverse field was important. Although for one position of this tube a field of 2 gauss caused a marked reduction in multiplier efficiency, for another position, fields of up to 8 gauss merely increased the efficiency by 20%. Therefore care was taken to position the 0° phototube according to the E.M.I. specification given in Figure 24.

(b) The tests already described in Section A3, in which the counting rate of the 90° scintillation counter was compared with that of the 0° solid state counter, would have exposed any dependence of counter efficiency on magnetic field. As these tests were carried out over the full range of particle energies, the 90° count was taken with many different values of magnetic field. The differences between the 0° and 90° counts were random and did not vary systematically with magnetic field strength. Hence the 90° counter was shown to be unaffected by the stray magnetic fields.
The $0^\circ$ counter was checked by counting a beam of neutral particles for various magnetic field strengths between 300 and 5000 gauss. The minimum of 300 gauss was necessary to remove all the charged particles from the $0^\circ$ detector. The neutral count was found to be constant to within 1% and showed no systematic variation with magnetic field.

4. Counting Statistics

The number of particles counted always exceeded $10^4$ in each counter. Hence, the percentage deviation, $\frac{100}{\sqrt{N}}$, was always less than 1%.

C. ENERGY MEASUREMENT

1. Magnetic Field

For internal comparison of the results obtained in these experiments, the accuracy of the energy measurements was entirely dependent on the accuracy of setting and reading the magnetic field. Experience showed that the magnetic field could be adjusted for peak counting rate to within $\pm$ 20 gauss at 2000 gauss and to within $\pm$ 50 gauss at 4000 gauss. As the energy was proportional to the square of the magnetic field, the percentage error in the energy was equal to twice that in the magnetic field. Therefore, for magnetic fields of both 2000 and 5000 gauss, maximum possible errors of $\pm$ 2% existed in the energies, due to uncertainties in the magnetic fields.

Hence for comparative studies of the results, the maximum possible error in the energy was 2%. However the absolute energy values included the uncertainty in the calibration of the spectrometer.
2. Calibration of Spectrometer

The formula for the conversion of the magnetic field readings (B Kilogauss) into energy values (E KeV) was

\[ E = 4.59 B^2 \]

Therefore, for an absolute energy scale, the error in the constant, 4.59, was added to the error due to the uncertainty in setting the magnetic field.

Thirteen measurements were made of B and V, from 0 to 120 KV, as described in Section E of Chapter 3. By the addition of 8.8 KeV to the value V, the particle energies were obtained and hence thirteen values of \( \frac{E}{B^2} \), with a mean value of 4.59. The root mean square deviation was calculated to be 0.7% of 4.59.

As a check, a least squares deviation line was fitted to the thirteen \((B^2, V)\) points. The slope of the line was 4.56 and at \( B^2 = 0 \), \( V = -8.5 \). This value of 8.5 KeV (i.e. the energy the particles gained from the D.C. converter voltages and the ion source) was only in fair agreement with the measured value of 8.8 KeV. However, it can be seen that this difference had very little effect on the constant of calibration, 4.56 and 4.59 agreeing to within 0.7%.

Therefore the constant 4.59 was used for conversion to absolute energy values, the probable error in this constant being less than 1%. Thus the maximum probable error in the energy measurements was 3%, the error in the magnetic field measurement being added to the error of calibration.
D. REPRODUCIBILITY OF MEASUREMENTS

As an ultimate check of the reliability of the apparatus, frequent measurements were made of the positive to neutral ratios of hydrogen particles emergent from the alumina backing foils before evaporation. The maximum variation recorded with measurements on a single batch of foils was $\pm 5\%$ and in general this was true for measurements on different series of foils (Figure 31). There were some measurements which deviated by almost twice the $\pm 5\%$ margin, but as the alumina foils were produced in aqueous solutions, it is not surprising that differences should have occurred in their surface composition. Therefore the agreement between charge ratio measurements on alumina foils produced from the same solutions demonstrated the reliability of the charge ratio measurements.

E. OVERALL ACCURACY

It has been shown that all significant sources of error in the measurement of the charge ratios are included in the test demonstrating to what degree of accuracy the total beam is equal to the sum of its components. The maximum variation in this test was found to be $\pm 2.5\%$. This included the statistical fluctuations in counting, which could amount to $\pm 1\%$.

In the measurement of energy there was shown to be:

a) A $\pm 1\%$ error in the comparative energy measurement, and
b) A $\pm 2\%$ error in the absolute energy measurements.

From a typical curve of $H^+/H_0$ plotted against energy, the $\pm 1\%$ error in comparative energy measurements can be shown to correspond to a $\pm 2\%$ error in charge ratio. Combined with the maximum variation above ($\pm 2.5\%$), a total error of about $\pm 5\%$ is obtained, which is in agreement with measurements of fluctuation on alumina foils.

Having established the reliability of the apparatus to this degree of accuracy, we can now examine the experimental results.
CHAPTER 5

RESULTS

INTRODUCTION

The apparatus described in the preceding chapters was used for three investigations of charge exchange effects in thin foils:

A. Measurement of the $\text{H}^+\!:\!\text{H}^0$ ratio.

If the small probability of double electron capture is neglected, the $\text{H}^+\!:\!\text{H}^0$ ratio is equal to the ratio of the cross sections for electron loss and capture by protons. Observations were made of the dependence of these cross section ratios on the energy of the hydrogen particles and on the characteristics of the material of the foil. These measurements, when combined with the data available on the cross sections in gases and dielectrics, yield information on the mechanism of the electron capture and loss processes in solids.

B. Measurement of the $\text{H}^+\!:\!\text{H}^0$ Ratio.

Similarly, the $\text{H}^-\!:\!\text{H}^0$ ratio is equal to the ratio of the cross sections for electron capture and loss by hydrogen atoms, double electron transfer again being neglected. Since it is not known how great an effect the double electron transfers have, and whether they can be neglected without introducing large errors into the deductions about cross sections, it is important that all three components should be measured.

C. Investigation of the Effect of Background Gas and Foil Structure on Charge Ratios.
Numbers give particle energies in Kev.

Figure 25

H⁺, H⁺ from ALUMINIUM
Present values

\[ \frac{H^+}{H^*} \]

Particle Energy KeV.

H^+/H* from COPPER

Present values

\[ \frac{H^+}{H^*} \]

Particle Energy KeV.

H^+/H* from GOLD

Figure 27
Present values

Phillips

Particle Energy Kev.

H⁺:H⁺ from SILVER

Figure 28
and 26 and the extrapolated values, corresponding to clean foils of aluminium, copper, silver and gold are plotted versus energy in Figures 27 and 28. The $H^+:H^0$ ratios for copper and silver varied in a way very similar to those of gold.

It can be seen from Figures 27 and 28 that the charge ratios from different foils of the same metal deviate from a smooth curve by more than could be explained by the experimental error of $\pm 5\%$. These variations were further investigated in an attempt to determine their origin. In particular, special precautions were taken to be sure that they did not arise from erratic behaviour of the accelerator or measuring equipment. The outcome of the investigations, which are presented and discussed in Section C, gives a clear indication that there are other important factors influencing the results.

In Figures 27 and 28 the extrapolated $H^+:H^0$ values for aluminium, silver and gold are compared with the only other reliable results available for these metals, those obtained by Phillips. The agreement can be seen to be good, the lines of best fit for the present results agreeing to within $5\%$ with the curves given by Phillips. In Figure 29 the results of the present investigations are compared with those of Phillips and of Hall.

In Figure 30 the $H^+:H^0$ values for the four metals, aluminium, copper, silver and gold, are plotted against particle energy. It is apparent from this diagram that the
COMPARISON OF $H^+H^0$ FOR PHILLIPS, HALL AND PRESENT VALUES

Figure 29
COMPARISON of $H^+\cdot H^*$ RATIOS from ALUMINIUM, COPPER, SILVER and GOLD

Figure 30
spread of results is too large to distinguish between copper, silver and gold. It is clear, however, that the H⁺:H⁰ ratio is considerably higher for aluminium than for the other metals.

ANALYSIS OF H⁺:H⁰ RESULTS

1. Electronic Properties of Target Atoms

When an attempt is made to account for the differences in the electron transfer processes in aluminium and the noble metals, copper, silver and gold, one might expect to obtain some correlation with the electronic properties of the metals. The relevant properties are listed in Table 1.

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
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</tr>
</thead>
<tbody>
<tr>
<td>z</td>
<td></td>
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<td></td>
</tr>
<tr>
<td>A1</td>
<td>13</td>
<td>2.7</td>
<td>6.02</td>
<td>72</td>
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<td>2.20</td>
</tr>
<tr>
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<td>111</td>
<td>5.5</td>
<td>4.41</td>
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<td>Au</td>
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<td>19.3</td>
<td>5.90</td>
<td>106</td>
<td>5.6</td>
<td>4.90</td>
</tr>
</tbody>
</table>

1 : z : atomic number
2 : ρ : density : gms cm⁻³
3 : d : number of atoms cm⁻³ x 10⁻²²
4 : V : binding energy of electrons in outermost filled shell : eV.
5 : E₄ : Fermi energy : eV.
6 : Ø : work function : eV.

A property in which the noble metals are similar and aluminium different is the first requirement for a
correlation with the $H^+:H^0$ ratios. It can be seen from Table 1 that neither the atomic number nor the density of the metal fulfil this criterion, for the magnitudes of these two properties form evenly increasing sequences. From column 3 it can be seen that the number of atoms per cubic centimetre also shows the wrong type of variation, as aluminium, silver and gold are similar and copper is the exception. Thus the density, the atomic number and the atomic density cast little light on the behaviour of the charge ratios.

The fourth column of Table 1 shows the binding energies of the most loosely bound electrons, apart from the valence electrons. For an electron to be captured from one of these levels it must gain at least 60 electron volts energy to transfer to the $-13.6$ volt ground state of hydrogen. The large energy change makes capture from these levels very unlikely. Therefore the exchange of electrons must take place between the valence states of the target atom and the bound states of hydrogen. In the metallic state, the valence electrons become the free conduction electrons, and as these are the electrons involved in charge exchange, we will examine some of their properties which seem relevant to the present enquiry. The properties of interest are:

a) Number of free electrons per atom.

b) Fermi energy.

c) Work function.

a) On first appearance, the number of free electrons per unit volume seems to be a significant property
for correlation, as aluminium has three times as many free electrons as the noble metals. However, any idea of a direct connection between the number of free electrons and the $H^+:H^0$ ratio has to be dismissed, because of the smaller, rather than larger, number of neutral particles emergent from aluminium. It is very difficult to see why an increased density of free electrons should cause a decrease in the number of neutralized protons.

b) The Fermi energy of the free electrons is shown in column 5 of Table 1, where aluminium can be seen to have a higher value than the other metals. Again however, the behaviour of the $H^+:H^0$ ratio deduced from the Fermi energies does not agree with the observed behaviour, for the effect of the free electron velocity would be greatest when it was equal to, or greater than, the proton velocity and the importance of the electron velocity would decrease at high proton velocities (58, 59, 63). The 11.7 eV electrons in aluminium have velocities equal to protons of 21.5 KeV. Therefore, if this electron velocity were to influence the charge ratio, it would do so at proton energies around 20 KeV and the charge ratios for different metals would converge, as the velocity of the hydrogen particles became large compared with the electron velocities. As can be seen from Figure 30, the observed behaviour of the $H^+:H^0$ ratio is precisely opposite to these deductions. Hence the Fermi energy also is not relevant to the question of charge ratios.
c) The work function can be seen to vary in the right way (column 6, Table 1), aluminium being substantially different from the noble metals. The correlation between work function and the proton content of the equilibrated beam has already been observed by Phillips (Figure 8), who saw this correlation as an indication that the charge ratios were changed as the particles emerged from the surface of the metal.

However, the fact that the correlation applies at high energies (>100 KeV) casts doubt on this hypothesis. For if the charge ratio characteristics were impressed upon the particles as they emerged from the surface, the correlation of charge ratio and work function would apply at low energies to a much greater extent than at high. This is because low-speed particles remain longer near the surface of the metal, allowing more time for the extraction of electrons.

Moreover, the work functions would be expected to affect the charge ratios in a way opposite to that observed. We would expect that the metal with the lowest work function, that is, the one with the most easily extracted electrons, would yield most neutral atoms. Since this is not so, the work function is also ruled out as an explanation of the low proportion of neutral atoms in aluminium.

We conclude, therefore, that the difference between the charge ratio of aluminium and the noble metals cannot be explained in terms of free electrons. This conclusion
Comparison of HH for Metals, Dielectrics and Gases

Figure 31
is supported by a comparison of the $H^+:H^0$ ratios for metals and other stopping media.

2. **Comparison with $H^+:H^0$ ratios from other stopping media**

When the equilibrium $H^+:H^0$ ratios for other media are compared with those of metals, they are found to be strikingly similar (see Figure 31). This similarity is found in dielectric foils, such as alumina in the present work and silicon monoxide as measured by Phillips (74).

Investigations have been reported (8, 85) for a number of gases, the ones shown being hydrogen, helium, nitrogen, oxygen, argon and neon. In all instances the results for gases have a close similarity to those for solid dielectrics and metals (Figure 31).

Therefore there is no reason to expect the charge exchange processes to vary greatly between metals and dielectrics, or solids and gases. As the charge exchange cross sections in gases are known to be determined by the wave functions of the loosely bound electrons in a complex way, it is reasonable to suppose that the same applies to solid target materials.

Thus we conclude that the differences between the charge ratios in aluminium and the noble metals are caused by differences in the wave functions of the outer electrons, and that the modification of the wave functions caused by the solid state does not have a large effect on the charge ratios.

Similarly, the difference between the Fermi energy in aluminium and in the noble metals is caused by
TYPICAL $H^- : H^0$ CURVES FOR ALUMINIUM

Figure 32
variations in the wave functions of the different metals. The same applies to the work function. Thus though there is a correlation between these two factors and the charge ratios, there is no direct connection. Rather it seems that all are determined, in a complex way, by the wave functions of the outer electrons.

B. MEASUREMENT OF $H^- : H^0$ RATIO

When measurements were made of the variation of the $H^- : H^0$ ratios with time, these were found to behave in a way very similar to the $H^+ : H^0$ ratios. That is, the $H^- : H^0$ ratios changed with time, moving towards the values yielded by contaminated surfaces.

As there was no additional data to be gained from further measurements of the time variation, the $H^- : H^0$ ratios were measured over the energy range from 20 to 80 KeV on single clean foils. The measurements of the $H^- : H^0$ versus energy curves were repeated at least six times for each metal, in order to check the reproducibility of the result. (Figure 32). The curves were consistent to $\pm 5\%$, with a few exceptions which deviated by up to $10\%$. These larger deviations were very similar to the large deviations found in the extrapolated $H^+ : H^0$ values. The most probable curve could be drawn for each metal, and these curves are shown in Figures 33 and 34. The $H^- : H^0$ against energy curves, obtained from the present work, are shown together in Figure 34. Here it is seen that the $H^- : H^0$ ratios for silver and gold are indistinguishable, but that, as in the case of the $H^+ : H^0$ ratios, aluminium yields
Present values
Phillips'
Fogel et al.

Particle Energy K.e.v.

$\frac{H^-}{H^*} %$

$H^-:H^*$ from ALUMINIUM

Figure 33

Particle Energy Kev.

$\frac{H^-}{H^*} %$

$H^-:H^*$ from SILVER

Figure 33
Comparison of $H^-:H^*$ Ratios from Aluminium, Copper, Silver, and Gold

Figure 34.
a higher proportion of charged particles. The behaviour of copper, on the other hand, is different from that observed in the $H^+:H^0$ experiment; the $H^-:H^0$ ratios for copper being closer to those of aluminium than to the other noble metals.

From Figures 33 and 34, it can be seen that for silver and aluminium the proportion of negative ions measured by Phillips (74) was smaller than in the present investigation, and that for gold, the curves of the two investigations intersect.

The only other measurements available for comparison were made by Fogel and his co-workers (31). In their measurements of $H^-:H^0$, at energies between 28.2 and 11.5 KeV, they found that copper and aluminium yielded very nearly the same ratios. With beryllium, aluminium and copper, their yields of negative ions were always greater than those of Phillips. An $H^-:H^0$ ratio for 28.2 KeV particles in aluminium has been calculated from a combination of their measurement of $H^-:H^+$ with the present measurement of $H^+:H^0$, for the same conditions of foil and energy. This ratio is shown in Figure 33, where it can be seen to be in better agreement with the present results than with Phillips' measurements.

As pointed out in Chapter 1, Fogel's measurements were made with little regard to foil contamination. However, from Figure 35 it can be seen that the variation between the results of the different workers is considerably larger than the changes due to contamination.
Phillips' results are unlikely to be as reliable for the measurement of negative ions as for the other two components. For the intensities of the $H^-$ components are only a few percent of the total beam, and his use of current measuring techniques would lead to serious errors if the detector were at all non-linear in its response. Although he claims to have tested for non-linearity, it is difficult to give an alternative explanation of the difference between his and the present results; for this difference is too large to be explained by an inadequate correction for the $H^0$ counting loss in the present investigations.

Phillips also states that the metals with the largest neutral component at low energies (in his case, gold and aluminium) also have the largest negative component. The present results disagree with this, for they show aluminium to have fewer neutral, and yet more negative, particles than the other metals. Moreover, Phillips' statement is not really very convincing, even within his own measurements. For the proportions of neutral particles he measured emergent from aluminium and gold are only 3\% or 4\% higher than from the other metals, and this applied only below 25 KeV. On the other hand he found the number of negative ions from aluminium and gold to be 20\% to 30\% higher than from the other metals, and this applied for energies up to 100 KeV. Thus there does not seem to be a direct connection between the numbers of neutral and negative particles.
ANALYSIS OF $\text{H}^-:\text{H}^0$ RESULTS

1. Comparison with $\text{H}^-:\text{H}^0$ ratios from other stopping media

   In Figure 36 it can be seen that both the $\text{H}^-:\text{H}^0$ ratios from alumina, measured in the present work, and the $\text{H}^-:\text{H}^0$ ratios from silicon monoxide, measured by Phillips, are very similar to those from metals. Hence we can conclude, remembering that the same was true in the case of the $\text{H}^+:\text{H}^0$ ratio, that charge ratios are not greatly influenced by the fact that the outer electrons are free in a metal and bound in a dielectric.

   On the other hand, the $\text{H}^-:\text{H}^0$ ratios for foils can be seen from Figure 36 to be higher than for gases. Thus, there appears to be a solid state effect, which causes a higher proportion of negative ions to be present in hydrogen particles equilibrated in solids, than in hydrogen particles equilibrated in gases.

2. Effect of Solid State on $\text{H}^-:\text{H}^0$ ratio

   An increase in the probability of double electron capture would explain the larger number of negative ions observed in solids. It seems that this increased probability must be caused by the far greater electron density, which would favour three body collisions in solids more than in gases.

   The cross sections for double electron transfer are often regarded as small, permitting the hydrogen system to be treated as a pair of two component systems. In the following calculation, the accuracy of this assumption is examined.
The six charge exchange cross sections are written in the form \( \sigma_{ij} \), the subscripts referring to initial and final states respectively. Thus we have, in the three charge states, the fractions \( F_i \), \( F_0 \) and \( F_T \) and the six cross sections \( \sigma_{io} \), \( \sigma_{ii} \), \( \sigma_{i0} \), \( \sigma_{iT} \), \( \sigma_{iT0} \) and \( \sigma_{iT1} \).

\( \pi \) is the distance travelled by the beam in the target, multiplied by the number of target atoms cm\(^{-3}\).

Then

\[
\frac{dF_i}{d\pi} = \sigma_{ii} F_i + \sigma_{i0} F_T - (\sigma_{i0} + \sigma_{iT}) F_i
\]

\[
\frac{dF_T}{d\pi} = \sigma_{iT} F_0 + \sigma_{iT} F_T - (\sigma_{iT0} + \sigma_{iT}) F_T
\]

and \( F_i + F_0 + F_T = 1 \).

Now, at equilibrium, \( \frac{dF_i}{d\pi} = \frac{dF_T}{d\pi} = 0 \).

Thus we can solve the three equations in \( F_i \), \( F_0 \), \( F_T \), to obtain the equilibrium ratio.

\[
\frac{H^-}{H^0} = \frac{F_T}{F_0} = \frac{\sigma_{iT} \sigma_{iT0} + \sigma_{iT1} (\sigma_{iT0} + \sigma_{iT})}{\sigma_{iT} \sigma_{iT0} + \sigma_{iT1} (\sigma_{iT0} + \sigma_{iT})}
\]

If double transfer is neglected the first term, in both numerator and denominator, is equal to zero. In this case we obtain the two component approximation.

\[
\frac{H^-}{H^0} = \frac{\sigma_{iT}}{\sigma_{iT0}}
\]

No comment on the validity of this approximation can be made on the basis of the values of the \( H^+:H^0 \) and \( H^-:H^0 \).
ratios, for we have six unknowns and only two equations.

The accuracy of the two component approximation can be tested for various gaseous targets in which the cross sections are known. The cross sections for 15 keV hydrogen particles in hydrogen gas (29, 30, 85) and 5 keV hydrogen particles in argon were substituted into both the exact and approximate formulae. The equilibrium fractions of negative ions are 2% and 3.6% respectively, in these two cases. In both, the value of the equilibrium ratio calculated by using the two component approximation, rather than the exact formula, was only about 3% too low.

As the $H^- : H^0$ ratios from solids are often 100% larger than the ratios in gases at the same energy, there must be a difference in at least one of the charge exchange cross sections. It seems that the double capture cross section is the one most likely to be increased by the condensation of the target atoms. If this cross section, $\sigma_T$, is about fifty times as large in a solid as in a gas, in a solid the $H^- : H^0$ ratio would be doubled, and the $H^+ : H^0$ ratio reduced by about 10%. This 10% change in the $H^+ : H^0$ ratios would not be observable, as it is small and would also most probably be masked by small changes in the other five charge exchange cross sections.

The hypothesis, that the solid state of the target caused the double capture cross section to be increased, is further supported by a comparison of the proportion of negative ions from the four metals. The order of decreasing
free electron density in the metals and of decreasing $H^-:H^0$ ratios are the same; namely aluminium, copper, silver and gold.

C. INVESTIGATION OF THE EFFECT OF BACKGROUND GAS AND FOIL STRUCTURE ON CHARGE RATIOS

As there are many extraneous factors which can influence the charge ratios emergent from evaporated foils, it is not possible to speak with authority about the measured charge ratios, unless these effects have been investigated. The factors which could affect the behaviour of the emergent ratios are:

1. Adsorbed layers of gas or oil on the foil.
2. Gas trapped in the foil during evaporation.
3. Thickness of foil.
4. Crystalline structure of foil.

1. Gas Adsorption

To study the effects of gas adsorption it is essential to know the thickness of material required to establish equilibrium. Phillips states that contaminating layers only a few atoms thick will completely alter the characteristic equilibrium ratio for the underlying solid. He also reports a measurement which showed the minimum thickness for equilibrium in aluminium to be six atomic layers.

It can be shown by calculation, later borne out by experiment, that these estimates of minimum thickness for equilibrium are too small.
Calculation of minimum thickness for equilibrium

For simplicity, a two component system will be treated. The fractions of the total beam in the two charge states are denoted by $F_0$, $F_1$, and we have

$$F_0 + F_1 = 1$$

We put $\sigma_{i0}$ and $\sigma_{o1}$ as the cross sections for electron capture and loss respectively.

We also put $\pi$ as the distance travelled by the beam in the target, multiplied by the number of target atoms cm$^{-3}$. This multiplier is assumed to be constant, the target being homogeneous in composition.

We can now write, for the growth of the neutral component as the beam traverses the target,

$$\frac{dF_0}{d\pi} = F_1 \sigma_{i0} - F_0 \sigma_{o1}$$

Substituting $F_1 = 1 - F_0$ we have

$$\frac{dF_0}{d\pi} = \sigma_{i0} - F_0 (\sigma_{i0} + \sigma_{o1})$$

The solution of this differential equation is

$$F_0 = \frac{\sigma_{i0}}{\sigma_{i0} + \sigma_{o1}} + B e^{-\left(\frac{\sigma_{i0} + \sigma_{o1}}{\sigma_{i0} + \sigma_{o1}}\right)}$$

Now $\frac{\sigma_{i0}}{\sigma_{i0} + \sigma_{o1}}$ is the equilibrium value of $F_0$, attained at $\pi = \infty$, and will be denoted by $F_{0\infty}$.

$B$ is a constant, dependent on the initial composition of the beam. If the initial value of $F_0$, at $\pi = 0$, is $F_{0i}$, then
Therefore \( F_0 = F_{0\infty} + (F_{0i} - F_{0\infty})e^{-(\sigma_0 + \sigma_t^e)\pi} \)

or \( \frac{F_0 - F_{0\infty}}{F_{0\infty}} = \frac{F_{0i} - F_{0\infty}}{F_{0\infty}} e^{-(\sigma_0 + \sigma_t^e)\pi} \)

This is our final result, giving the fractional deviation from equilibrium, in terms of the initial deviation, the distance travelled in the target, the target density and the cross sections for the charge changing collisions.

We can now make the condition for equilibrium that \( F_0 \) should be within \( 1\% \) of \( F_{0\infty} \) and calculate two cases. In the first, we will put the initial fraction \( F_{0i} \) at a value differing from the equilibrium fraction \( F_{0\infty} \) by \( 10\% \), and in the second \( F_{0i} = 0 \); that is, the beam is entirely positive.

**Case 1.**

\[ 0.01 = 0.1e^{-(\sigma_0 + \sigma_t^e)\pi} \]

\[ \pi = -\frac{\ln 0.1}{\sigma_0 + \sigma_t^e} \]

\[ = \frac{2.3}{\sigma_0 + \sigma_t^e} \]

Inspection of previously measured values of the capture and loss cross sections in gases (1) shows that the sum of the two is in general between 5 and \( 1 \times 10^{-16} \) cm\(^2\) per atom. For this order of magnitude calculation, we will take the mean value of \( 2.5 \times 10^{-16} \) cm\(^2\) per atom as the sum of the cross sections for both metals and condensed gases and vapours.

Substituting this value for the cross sections,
we have

\[ \pi = 10^{16} \text{ atoms cm}^{-2} \]

The atomic density for aluminium, gold and silver is \(6 \times 10^{22}\) atoms cm\(^{-3}\).

Therefore the thickness required for equilibrium is

\[ T_e = \frac{10^{16}}{6 \times 10^{22}} \]
\[ = 3 \times 10^{-7} \]
\[ = 30 \, \text{Å} \]

**Case 2.**

If \(F_i = 0\), it is easily seen that the thickness required for equilibrium is double this value; that is, 60 Å is required for equilibrium.

If the value of the interatomic spacing of the metals, 2.5 Å, is combined with the calculated value of equilibrium thickness, the number of atomic layers necessary for equilibrium from a 10\% deviation is found to be

\[ N_e = \frac{30}{2.5} \]
\[ = 12 \text{ atomic layers}. \]

Thus, for the charge ratios to be characteristic of the adsorbed gas, a layer twelve atoms thick would be required.

It is unlikely that adsorption of gas would build up a layer as thick as this, unless there were a large amount of organic vapour in the system, which could condense on the foil surface.
FIELD IN MAGNETIC SPECTROMETER — GAUSS

$H^+ / H^o$ FOR ALUMINA AND VACUUM-AGED ALUMINUM
OLD FOILS
Alumina, Gold & Silver
Copper & Phillips'
Aluminium

NEW FOILS
Aluminium
Copper, Silver & Gold

\[ \frac{H^*}{H^*} \text{ from VACUUM AGED FOILS} \]

Figure 38
The calculation shows that the importance of gas adsorption has perhaps been overestimated; the three experimental investigations which follow support this view.

i) Vacuum aged Foils

The first experiments made on the effects of gas adsorption were those carried out on foils, which had been evaporated in the target chamber in the normal way, but were then allowed time for some surface contamination to take place. It was found that the charge ratios came to steady values about 30 minutes after evaporation. It was concluded that a certain amount of gas adsorption took place on the clean foil, but that the rate of adsorption after 30 minutes became very small. However, a further change took place when the foils were left in the target chamber overnight when the zeolite cold trap was outgassed. The outgassing liberated large quantities of gas, causing further condensation on the surface of the foil.

Curves from these contaminated, vacuum aged foils are shown in Figures 37 and 38. The measurements for copper were made on foils that had aged in the vacuum for a few hours, whereas the curves for the other metals were made after the foils had aged overnight. The effect of the extra condensed material can be seen by comparing the results of the relatively uncontaminated copper with those of silver and gold. A few measurements were taken which indicated that copper yielded the same charge ratios as silver and gold after overnight ageing. However, it can be seen from Figure 38 that,
even after overnight ageing, the characteristically high $E^{+}:H^0$ ratio of aluminium was observable through the adsorbed layers of gas or oil. If the thickness of these adsorbed layers had been sufficient to cause equilibrium, all the contaminated foils would have yielded the same $E^{+}:H^0$ ratio. As the ratio for old aluminium is different, it is clear that the thickness of adsorbed gas was never sufficient to cause equilibrium.

These results are in conflict with Phillips' observation that all foils give the same equilibrium charge ratio when contaminated. His curve for "dirty surfaces" happens to coincide with that for copper in Figure 38, but exact comparison of results from heavily contaminated foils is not fruitful, as the nature of the adsorbed material is variable. On the other hand, the present results are in agreement with those of other workers who have measured differences between contaminated foils. Hall, for instance, detected charge ratio differences between contaminated gold foils on the one hand, and oxidized and contaminated foils of aluminium, beryllium and silver, on the other. Similarly, Fogel et al. (31), in their measurements of the $H^-:H^+$ ratio, detected differences between contaminated foils of copper, beryllium and aluminium. For comparison with the present results see Figures 29 and 35.

ii) Time variation of $H^+:H^0$ from new foils

From Figures 25 and 26 which show the time variation of the $H^+:H^0$ ratios, it can be seen that there was
considerable variation in the rate at which the charge ratios changed. Now, the rate of adsorption on a clean surface varies directly as the gas pressure. Hence, if the charge ratios had changed with time, due to the adsorption of gas, the fastest changing ratios would have corresponded to the highest pressures in the target chamber. However, as can be seen from the table below, there was no correlation between pressure and the rates of change of the charge ratios. Therefore it must be concluded that the changes were not primarily due to the adsorption of gas.

<table>
<thead>
<tr>
<th>S</th>
<th>Pe</th>
<th>Pm</th>
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<td>5</td>
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</tr>
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<tr>
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<td>8</td>
</tr>
<tr>
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<td>10</td>
<td>5</td>
<td>1.8</td>
<td>10</td>
<td>6</td>
</tr>
</tbody>
</table>

- Table 2 -

S = slope of \( \text{H}^+:\text{H}^0 \) versus time line.
Pe = pressure in \( 10^{-7} \) torr during evaporation.
Pm = pressure in \( 10^{-7} \) torr during measurement.

The measurements of the rate of change of the \( \text{H}^+:\text{H}^0 \) ratio provide further evidence supporting the view that
gas adsorption is not the most important factor determining the change of ratio. From Figures 25 and 26 it can be seen that there is no very great difference between the rates of variation of the charge ratios from aluminium and gold. As aluminium is more reactive chemically than gold, a greater difference between the rates of change of the charge ratios for the two metals would have been seen if the change were due to gas adsorption; for adsorption would necessarily entail some oxidation and perhaps other chemical reactions.

That the results are independent of target chamber pressure is again evident when we consider the magnitude of the extrapolated $H^+:H^0$ values. For there is no correlation between the target chamber pressure and the magnitude and direction of deviation of the extrapolated values from the most probable $H^+:H^0$ versus energy curve.

Thus there is much experimental evidence in the present investigation supporting the view that the change in the charge ratios with time was not mainly due to the adsorption of gas. Again these results differ from Phillips' measurements, in which he observed a direct relation between the rate of change of charge ratio and the pressure (Figure 6a). By varying the gas pressure between $4 \times 10^{-6}$ and $2.4 \times 10^{-5}$ torr, he caused a corresponding change in the charge ratios. He also found a far greater difference between the rates of change of charge ratio for aluminium and gold than was found in the present investigation (Figure 6b). The faster change in the charge ratio from aluminium he attributed
to the fact that it oxidized, as well as adsorbing gas.

The effect of gas adsorption has been shown to be larger in Phillips' investigation than in the present work. This is undoubtedly due to his inferior vacuum and perhaps to the condensable nature of the residual gas. His experiments were carried out at a pressure of $5 \times 10^{-6}$ torr, which was at least five times higher than the pressure used in the present investigation. It is also likely that the conventional cold trap he used was not as effective for absorbing organic vapours as was the zeolite cold trap. Organic molecules condense very readily on the surface of a foil, and the thick layer of contamination so formed would exert considerable influence on emergent charge ratios. These factors could well have led Phillips to overlook the effects of foil structure which will be described in Part 4 of this section.

iii) Heated Foils

An attempt to reduce the rate of gas adsorption, and hence the rate of change of the charge ratios, was made by evaporating films onto heated backing foils. The alumina backing foils were heated to about $300^\circ$C before evaporation of the films and kept at this temperature during measurements. It was expected that the charge ratios would change more slowly, because of the decreased probability of incident gas sticking to the surface of the hot foils (17, 40). It was realised that the heating of the backing foils would also affect the crystallization of the evaporated layer and that
this crystallization might mask any surface changes.

A typical result of the measurements on heated foils, showing the variation of $\text{H}^+:\text{H}^0$ ratio with time, is given in Figure 26. The $\text{H}^+:\text{H}^0$ ratio is seen to change more quickly and to attain a steady value sooner for hot foils than for cold. This behaviour indicates that the change in charge ratio was not due to gas adsorption, for, if it were, the heated foils would yield a linear time variation at a reduced rate of change.

The shape of the curve, showing the variation with time of the $\text{H}^+:\text{H}^0$ ratio from hot foils, suggests that the change in the charge ratio is primarily due to a process involving movement of the atoms of the evaporated metal, such as crystallization or aggregation. For heat would hasten such processes and the condition for steady charge ratios would be achieved more quickly.

2. **Gas Trapped in Foils**

It is possible for large amounts of gas to be trapped in vacuum evaporated metal foils. The factors which influence the amount of trapped gas are, the pressure during evaporation, the rate of evaporation and the sticking probability of the gas atoms on the clean metal.

To quote an example, it has been calculated (20) that a tin film evaporated at a rate of $10^6$ Å per second in a vacuum of $10^{-6}$ torr will contain 1% gas atoms if the sticking probability is 0.1. However, if the sticking probability were 1.0, as could be the case for a reactive metal like aluminium,
the gas trapped would amount to 10% of the atoms in the foil.

To test for a connection between the amount of gas trapped in the foil and the charge ratio, a series of aluminium and gold foils were evaporated at different rates and at different pressures. The vacuum was varied between $8 \times 10^{-7}$ and $3 \times 10^{-6}$ torr and the evaporation rate between 1 and 100 Å per second. No connection was detected between these two parameters and the behaviour of the charge ratio. Therefore it was concluded that, under the conditions of the experiment, the gas trapped in the evaporated metal layer had very little effect on the charge ratios.

3. **Thickness of Foils**

As the thickness of the evaporated films varied considerably from foil to foil, it was easy to look for a connection between the behaviour of the charge ratios and the thickness of metal traversed by the hydrogen particles. Measurement of the thickness of the evaporated metal was effected by making proton stopping power measurements on the foils at 56.2 KeV. The stopping power of the alumina foils, which was consistently found to be 4.2 KeV at this energy, was subtracted from the total, to give the thickness of evaporated metal. The stopping powers of the metal films were found to vary between 1 and 5 KeV which corresponds to a thickness between 70 and 500 Å. Thicker films than these were not used because of the resulting excessive scattering and energy straggle.
Electron Micrographs of Silver Foils (×55,000)
Figure 39.
Electron Micrographs of Silver Foils (X55,000)

Figure 40
No direct connection between these thickness measurements and the behaviour of the $H^+:H^0$ ratio was found. In view of the calculation that about 30 Å of metal is sufficient for charge equilibrium to be established, this result is not unexpected.

4. **Crystalline structure of the foils**

Investigation of the structure of the foils proved to be difficult, because of the very small size of the crystallites. Various foils examined under a microscope appeared bright and even, but it was known that the appearance of evaporated films is often misleading. Moreover, the experiments with heated foils had indicated that the crystalline effects might be important.

It was thought, therefore, that important information on the foil structure could be revealed through examination by electron microscope. Towards the end of the experimental work, there was an opportunity of using the electron microscope of a neighbouring department, a model JEM-T6 made by the Japan Electron Optics Laboratory. A selection from foils of the four metals was examined. In general, the foils were too thick for much detail to be seen, as they consisted of at least two layers of evaporated metal. In order to overcome this difficulty, the foils for one set of measurements, the $H^-:H^0$ ratio of silver, were used for one evaporation only. An electron micrograph and an electron diffraction pattern were prepared from each foil (Figures 39, 40, 41 and 42).
Electron Micrograph of Silver Foil deposited on Hot Alumina (X55,000)

This picture is included, even though it is slightly out of focus, as it was the best obtained from a heated foil.

Figure 41
Electron Diffraction Patterns from Silver Foils

Figure 42
The electron diffraction patterns, with one exception, were very similar to the typical pattern shown in Figure 42a. The ring patterns revealed that the foils consisted of small crystallites, oriented at random. The only deviation from this standard diffraction pattern was obtained from a foil which had been deposited on a heated substrate. With this foil, Laue spot patterns were obtained very easily, indicating that the crystal size was considerably larger than that in the normal foils (Figure 42b).

From this evidence, it was deduced that the crystal size did not cause a significant variation in the charge ratios. For the heated foils, with their larger crystal size, yielded the same charge ratios as the normal foils, within the present limits of error. Even if equilibrium charge ratios did vary with crystal structure, the random orientation of the crystallites would have caused the effect to average out over the area of foil through which the beam passed. The area covered by the proton beam (1 mm. diameter) was one thousand times larger than the area of foil shown in each photograph. Thus there is little possibility that the measured charge ratios were affected by channelling of the particles along crystal planes in a way similar to that observed in stopping power measurements (64).

On the other hand, the electron micrographs reveal such a high degree of aggregation, that it is clear that the measured charge ratios must have been affected by the non-uniformity of the foils. The charge ratios emergent
Figure 43

H/H₀ for silver foils shown in electron micrographs

Field in magnetic spectrometer - Gauss
from these foils would have been a weighted mean of the ratios characteristic of the freshly evaporated metal and the material of the substrate.

This was borne out by a comparison of the electron micrographs with the corresponding $H^+;H^0$ curves obtained for each of the silver foils (Figure 43). The comparison revealed a correlation between the proportion of surface covered by silver and the corresponding $H^+;H^0$ ratio. The ratio tended to be higher for those foils in which the largest area of backing foil was exposed; this tendency agreed with the fact that alumina gave a higher $H^+;H^0$ ratio than silver.

The non-uniform foil thickness was also revealed in the energy straggle, though not very clearly, because of the low resolution of the magnetic spectrometer. The profiles of the primary proton beam and of the beam emergent from a silver foil are shown in Figure 23. Assuming that the profiles are Gaussian, we can calculate the standard deviations of the proton energies in these two beams by multiplying the full width at half height by 0.42. The standard deviation of the primary beam is given by $\sigma_1 = 0.42 \times 2.3 = 0.96$ KeV and that of the emergent beam by $\sigma_2 = 0.42 \times 3.4 = 1.6$ KeV.

The standard deviation caused by straggling has been measured (55) at 800 KeV and found to be 8.8 KeV mg$^{-1}$ cm$^2$, which is in agreement with theory (13). Since at 50 KeV, the standard deviation due to straggle would be less than this, the above value gives an upper limit to the straggle. The
standard deviation representing the effect of straggle in the 10 mg cm\(^{-2}\) silver foil is then 0.88 KeV (\(\Omega\)). The standard deviation in the emergent beam is given by

\[
\Omega_2^2 = \Omega_1^2 + \Omega^2
\]

\[
= (0.96)^2 + (0.88)^2
\]

\[
= 0.92 + 0.77
\]

\[
= 1.69
\]

\[
\therefore \Omega = 1.3 \text{ KeV}
\]

This expected value for the emergent beam is 80\% of the standard deviation measured (1.6 KeV).

The difference between these two values would be greater if a smaller detector had been used in the magnetic spectrometer. For the sensitive area of the solid state detector was too large to measure correctly the small energy spread of the primary beam.

Within the range of the present experiments, the rate of evaporation and the thickness of the foil did not affect aggregation. If they had, variation of thickness, or of evaporation rate, would have yielded systematically varying charge ratios.

It has been shown that the scatter of measured charge ratios was caused by different degrees of aggregation of the evaporated metal films. The charge ratios changed with time in different ways, depending on the rate of aggregation of the films during the process of measurement. It was the changing foil structure which had most influence.
on the ageing foils, not the adsorption of gas or oil.

It is interesting to note that Phillips did not observe the same sort of charge ratio variation as was found in the present work. Thus it is implied that his foils did not suffer from aggregation. His measurement of the minimum thickness of evaporated metal necessary for equilibrium, provides further evidence to support the implication. He found the minimum thickness to be only 50 eV. With this extremely thin layer, complete coverage of the backing foil would have been impossible, if the aggregation had been as severe as that shown in the electron micrographs of the silver foils. Although he used evaporated layers at least 250 eV thick, his foils were still very thin compared with those of about 2 KeV used in the current investigation.

It is possible that some aggregation did occur in Phillips' foils, probably to a lesser extent than reported in this work, but remained unnoticed because of the masking effect of gas adsorption. (See Part 1 of this section). The likelihood of Phillips overlooking the effect of aggregation was increased by his lack of attention to the problems of foil structure. On the other hand it seems that several experimental factors may have combined, by chance, to make his foils laminar.

The factors are concerned with the nature of the backing foils he used. These were made by evaporating aluminium on to a detergent-covered, glass plate and later floating the thin film off in water. The foils had stopping
powers of 4-7 keV and, although it is not reported, were presumably found to be free of pinholes. Because of their uncertain structure, brought about by vacuum evaporation, they would seem to be inferior to the alumina foils used in the present research. However, the aluminium backing foils may have had the advantage of alloying slightly with the first few layers of evaporated metal, thus preventing aggregation (89).

An alternative explanation of the smoothness of the metal films involves the adsorbed gas on the backing foil. The relatively high gas pressure in Phillips' apparatus could have helped an even deposition of metal, for evaporated atoms have considerably less mobility on gas covered surfaces. From this point of view, electron bombardment of the alumina foils carried out in the present investigations might well have encouraged aggregation of the evaporated metal.
CHAPTER 6

CONCLUSION

From the analysis of the experimental results in the previous chapter, we have drawn a number of conclusions.

1. When a beam of hydrogen particles passes through a foil with energies above 20 KeV, the equilibrium charge ratios are determined principally in the interior of the foil and not at its surface.

   This conclusion was drawn when it had been shown by experiment that there is no correlation between the $H^+:H^0$ ratios emergent from metal foils and the work functions of the metals. Corroborative evidence was provided by the observation that there is practically no difference between the $H^+:H^0$ ratios in metallic foils, dielectric foils and gases. This implies that the charge ratios are determined by the atoms themselves and not by a surface property.

2. The only charge exchange process which depends on the phase of the target material is one which produces a higher proportion of negative ions in the condensed phases.

   It was found that metals and solid dielectrics yielded very similar equilibrium charge ratios, and that these differed from the ratio in gases only by an increase in the relatively unimportant negative ion component. Thus, although no work has been carried out with liquid targets, we can surmise that the charge changing processes are almost independent of the phase of the stopping medium.
The observation of larger proportions of negative ions emergent from the solid targets led to the hypothesis that the greater electron density in solids brings about an increase in the probability of double electron capture. While the present investigation confirmed the previously measured proportions of the positive and neutral components to within the experimental error of ±5%, there was a significant difference in the proportion of negative ions measured. This difference was at times as high as 100%.

The present results for negative ions are to be preferred to the earlier ones. The use of particle counts instead of current measurements recorded on a chart led to greater accuracy. As the proportion of negative ions is always low, their measurement entails the comparison of currents of widely differing intensity. The linearity of particle counters is a valuable asset in the task of performing this comparison.

The adsorption of gas on the surface of the foils can have a considerable effect on the emergent charge ratios. However, there has been a tendency to overestimate the importance of this effect, for it was found that an oil diffusion pump and a carefully designed cold trap can achieve a vacuum adequate for the measurement of charge ratios characteristic of the material of evaporated foils.

The aggregation of the evaporated metal into separate clusters of crystallites is an important experimental factor in the measurement of charge exchange in thin foils.
Although a great deal of work has been carried out on the structure of thin films, the results of this work have been almost completely overlooked in the field of atomic collisions. The foils in all previous atomic collision experiments have been assumed to be very nearly laminar, whereas this is in fact usually far from being true. There is a need for further investigations of methods for producing laminar evaporated films.

There are many further experiments that could be performed to cast more light on the problems of charge transfer processes. For example, it would be interesting to attempt to measure the charge ratios emergent from monocrystalline foils. These foils would have to be evaporated on a hot backing foil and there could be considerable difficulty in keeping the evaporated film flat. Another useful investigation would be the measurement of the charge ratios of beams reflected from solids. This could well be a more suitable technique for the measurement first suggested, because of the ease of handling a solid single crystal instead of a foil. However, it would have to be ascertained that the particles penetrated the crystal sufficiently for the interaction to take place inside the solid rather than at its surface. It could also be illuminating to measure the charge exchange cross sections and equilibrium ratios in metals in the vapour phase. These cross sections should yield a clear indication of any differences between solids and gases as
charge equilibrating media.
production of backing foils:

1. Plastic Backing Foils

The plastic backing foils were made in a way similar to that described by Pate & Yaffe (72).

To make these foils, a co-polymer of 90% polyvinyl chloride and 10% polyvinyl acetate was dissolved in 3 parts by volume of cyclohexanone. The powdered co-polymer dissolved slowly, forming a clear solution. A drop of this solution was put onto the side of a dish of distilled water and allowed to trickle down into the water. As soon as the drop of plastic solution touched the water, a film spread over the surface. The end of the film furthest from the drop of co-polymer solution was then lifted with a glass rod and drawn out so that the drop continued to feed a thin layer onto the water surface.

Small aluminium discs (⁵⁄₈" diameter, 0.010" thick) with 3/16" holes in the centres were dropped onto parts of the film. The outside edges of the discs were next separated from the rest of the film by means of a glass rod dipped into cyclohexanone. Each disc was then lifted gently from the water.

The aluminium discs were designed to be clamped to the cups of the foil-carrying wheel, and in this way the plastic foils were held in the target chamber.

2. Alumina Foils

The following technique for the preparation of
alumina foils is a modification of two published methods (39, 41).

The same type of aluminium discs as described in (1) though without the central hole, were used for making these foils. Use of this standard size disc for both types of foil permitted either to be mounted in the cups without difficulty. The discs used for the preparation of the alumina foils were punched from aluminium sheet which had been buffed and polished on one side.

The discs were cleaned by immersion in a boiling solution of sodium carbonate and rinsing in distilled water. They were then anodized in a solution of equal parts of tartaric acid and ammonium tartrate, the concentration of the solution being unimportant. Another piece of aluminium was used as the cathode and the anodizing voltage was kept as low as possible, in order to produce thin foils. The anodization was carried out at 25 volts, for with lower voltages the foils were found to be too fragile to be very practical. After a few minutes in the anodizing bath, the discs were removed, rinsed and then placed in a clamp as shown in Figure 44, with the polished face of the aluminium downmost. A drop of hydrofluoric acid was used to remove the oxide layer from the centre of the top face. After this acid was washed away, some dilute hydrochloric acid, with a small amount of copper chloride added, was put into the well above the aluminium disc. The acid dissolved the aluminium, leaving the bottom oxide layer stretched across the hole.
The progress of the aluminium dissolving was observed by placing the clamp on a sheet of glass illuminated from below. When the light could be seen clearly through the acid and alumina, the acid was removed and the clamp opened. The remaining acid was washed away by holding the disc edgewise under a gentle stream of tap water. After drying, the foils were ready to mount. Some of the foils broke on drying, but the overall loss throughout the whole process was less than 20%.

Great care was necessary in the initial pumping down of the vacuum, when these alumina foils were used. To avoid breaking the rather brittle foils, it was found best to begin pumping through a tightly clamped rubber hose, which was very slowly released. Thus the number of foils broken during pumpdown was kept very low.

These alumina foils were thought to be more suitable than those that could be made by evaporation onto a solid substrate and then stripped off. (24, 81). A review of the various types of target foils used has been made by Yaffe (94).
CLAMP FOR ALUMINA FOILS

10 KV. QC. CONVERTER

Figure 44
There are many published works on the design of transistor D.C. converters (18, 19, 46, 86, 93, 95). The design used, which was based on a circuit given by Light & Hooker (53), is shown in Figure 44.

A voltage stabilizing circuit has been added and some modifications made to the published design to suit the available transistors.

<table>
<thead>
<tr>
<th>Input</th>
<th>12 volts D.C. 200 m.a.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Output</td>
<td>10 KV D.C. 100 μ.a.</td>
</tr>
<tr>
<td>Oscillating frequency</td>
<td>1.5 K c/s</td>
</tr>
<tr>
<td>Transistor, Power</td>
<td>0.0.26</td>
</tr>
<tr>
<td>Feedback</td>
<td>0.0.76</td>
</tr>
<tr>
<td>Rectifiers</td>
<td>F M I L 6 (International Rectifier Company)</td>
</tr>
<tr>
<td>Transformer</td>
<td>Wound on Ferroxcube U and 1 core (FX.1036, FX.1067) with 1 mm air gap</td>
</tr>
<tr>
<td>Primary</td>
<td>2 layers of 23 s.w.g. of 25 turns each in parallel.</td>
</tr>
<tr>
<td>Secondary</td>
<td>5000 turns of 45 s.w.g.</td>
</tr>
<tr>
<td>Base</td>
<td>15 turns of 32 s.w.g.</td>
</tr>
<tr>
<td>Feedback</td>
<td>45 turns of 32 s.w.g.</td>
</tr>
</tbody>
</table>
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