N94- 30485

A FUTURE, INTENSE SOURCE OF NEGATIVE HYDROGEN IONS

Hugh Siefken
Greenville College Department of Physics
Greenville, IL, 62246

Charles Stein
Phillips Laboratory, Kirtland Air Force Base
Albuquerque, NM 87117

ABSTRACT

By directly heating lithium hydride in a vacuum, up to 18 μA/cm^2 of negative hydrogen has been obtained from the crystal lattice. The amount of ion current extracted and analyzed is closely related to the temperature of the sample and to the rate at which the temperature is changed. The ion current appears to be emission limited and saturates with extraction voltage. For a fixed extraction voltage, the ion current could be maximized by placing a grid between the sample surface and the extraction electrode. Electrons accompanying the negative ions were removed by a magnetic trap. A Wein velocity filter was designed and built to provide definitive mass analysis of the extracted ion species. This technique when applied to other alkali hydrides may produce even higher intensity beams possessing low values of emittance.

INTRODUCTION

As our high level of technology continues into the future, the basic assumption made by everyone is that sufficient electrical energy will be available. The method of producing the energy is the uncertain factor. The procedures[1] used to initially estimate future fossil fuel supplies have been remarkably accurate especially when applied to national resources of oil and natural gas. The use of coal has seen increased growth during the past decade but at a price. This is because the total cost of burning coal for the long term includes the damaging effects of the effluents on materials and the possible effects of loading the atmosphere with CO_2.

An alternative is the process of nuclear fusion. This is a process which requires a machine to hold the reactants together long enough, and close enough, for the fusion of two nuclei into a product nucleus. One such machine is the TFTR at Princeton. The reactants are held within a strong magnetic field away from the vacuum chamber walls so losses are minimized. To supply the heat energy for the plasma at the place where it is needed, fast neutral atoms are used to penetrate the magnetic field which surrounds the reaction volume. In the TFTR positive ions are generated and then neutralized. However for the next

411

generation of Tokamak machines, planning is centered on the use of negative ions which can be more efficiently neutralized than the fast moving positive ion. Consequently, high current and high energy negative hydrogen (deuterium) ion beams are considered one of the best ways to achieve plasma heating and to provide for plasma diagnostics in these future machines[2,3]. The negative ion can be accelerated to the desired energy (150 - 500 keV) and passed through a gas which removes the weakly bound (~ .75 eV) electron resulting with neutral hydrogen which can penetrate the magnetic field used for confinement.

During the last decade, the development of ion sources has seen remarkable progress. Surface ionization, sputtering, and volume ion sources have each been developed so that the merits and drawbacks of each type are generally known. In some cases the precise physical mechanism for the ion production is not totally clear but enough is known in each of these cases to make a viable ion source for the respective application.

Figure 1 shows a schematic of a volume H^- ion source which has been developed at Lawrence Berkeley Laboratory[4]. It is representative of a larger group of H^- ion sources of this type produced in several laboratories. This ion source is attractive because it produces H^- at a low ion temperature (~ .3 eV) which results in lower beam emittance than some other types. However, the tungsten or LaB_6 filaments have a finite lifetime that eventually terminate due to erosion or fatigue. Also an arrangement of strong permanent magnets must be used to confine the plasma which is needed for the eventual production of the H^-.

A variation of this is to replace the filament with an R-F discharge antenna. This produces the plasma effectively but typically a 50 kW power supply with a 2 MHz oscillator is required to produce the ions.

Various geometrical arrangements and operating conditions have increased the current obtainable from these sources, but they all require the production of a dense plasma with which is associated corresponding difficulties in maintaining the critical conditions required for optimum performance, especially in the pulsed, high arc current mode of operation.

In contrast to these present day H^- sources I now would like to present some of the initial work done at the Phillips Laboratory in which H^- was produced by a more elementary method. This method involves the formation of H^- ions simply by heating a compound, LiH, which is an alkali metal hydride. There are two important factors which converge to make this class of compounds the basis for a possible intense H^- (D^-) source.
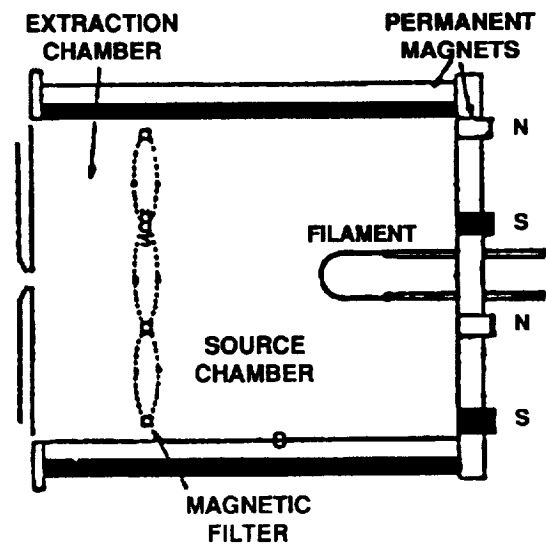

Figure 1. Schematic of LBL multicusp volume source. From Ref [4].

First, they store large volumes of hydrogen. It is been known for some time that metal hydrides permit the storage of hydrogen more efficiently with respect to volume than even liquid or solid hydrogen. Table 1 indicates some relative storage capacities of hydrogen.

Table 1. HYDROGEN DENSITIES IN SOME HYDROGEN-CONTAINING COMPOUNDS. FROM REFERENCE [5]

Compound	Number of Hydrogen Atoms per cm^3 X 10^{-22}
Liquid hydrogen (20°K)	4.2
Solid hydrogen (4.2°K)	5.3
LiH	5.9
TiH_2	9.2
ZrH_2	7.3
YH_2	5.7
UH_3	8.2

Formation of the hydride occurs when a given amount of hydrogen gas at a fixed temperature is brought into contact with the alkali metal. Some of the hydrogen is absorbed by the metal until a pressure equilibrium is reached at which time the hydrogen pressure becomes fixed. If more hydrogen is added, more is taken up by the metal until a new equilibrium pressure is reached. Thus for a given temperature an equilibrium pressure of hydrogen is reached. To reverse the hydrogen storage process the solid hydride is heated. As the temperature is increased higher equilibrium pressures of hydrogen are released. For LiH at 700° the equilibrium pressure is 28 mm.[6]

The second factor which makes this class of compounds feasible as a future source of negative ions is the strong ionic bonding between the constituent atoms. As hydrogen is taken into the metal, the metal lattice spacing decreases due to the strong attraction between the metal cation and hydrogen anion. The density of the hydride is thus much larger than the metal. The relatively large crystal lattice energies, defined as $MH \rightarrow M^+ + H^-$, vary from 9.5 eV for LiH to 6.7 eV for CsH. Various experiments using LiH confirms the presence of a bond which is strongly ionic. Calder[7] et al., using x-ray measurements, estimated the nature of the LiH bond to be between 80% and 100% ionic. Using electronegativity methods, Pretzel et al.[8] estimated the LiH bond to be 87.5% ionic. Additional support is given to this ionic bond model by the fact that when molten LiH is electrolyzed, hydrogen appears at the positive electrode[6] and the observation by Palmer[9] of the breaking of LiH into H^- when impacted on heated tungsten.

EXPERIMENTAL SET-UP AND RESULTS

Due to the relative ease of handling compared to some of the other alkali metal hydrides we chose to use LiH. The experimental arrangement used to measure the electron and ion currents as the temperature of the LiH was changed is shown in Figure 2.

A small piece of solid LiH, mass ~ 0.4 g, was removed from a large block stored in a tight drum, crushed, put in the sample container of the apparatus, and immediately loaded into the vacuum system. Any small amount of water vapor absorbed from the low humidity of the environment or remaining from the manufacturing process was found to be released in the vacuum system several minutes prior to the production of H^-. A residual gas analyzer was used to monitor the presence of water vapor and other gases present in the vacuum system during each run as the temperature of the sample was increased. The crushed LiH was held in a stainless-steel container

Figure 2. Experimental circuit used to measure electron and ion currents.

which also contained an iron-constantan thermocouple and tungsten heater. The heater and thermocouple were floated at the same (acceleration) potential as the sample holder. Electron and ion currents were measured with respect to ground potential.
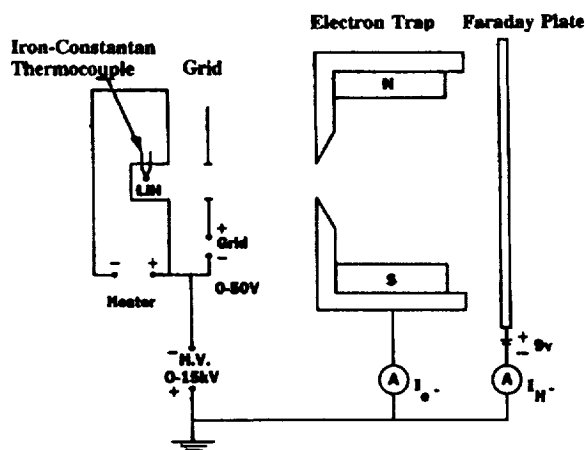
To separate the electrons from the negative ions an electron trap with a magnetic rigidity $\int Bdl$ = 3000 G cm and average field of 600 G was used. A Faraday plate made of carbon to minimize the production of secondary electrons was used to collect the ions. A small positive bias voltage of 9 V on the plate made approximately an 8% effect on the magnitude of the H^- current by forcing secondaries back to the carbon plate.

Three different heater geometries were used. A flat heater, a helical coil, and a boat-shaped heater were each used to heat the sample. The results of heating a larger volume with a boat and helical coil did produce more stable currents but did not produce larger currents than the flat heater.

Hydrogen was removed from the sample by heating the sample over a range of temperatures up to 800°C. At a given temperature the hydrogen is removed from the sample at an equilibrium pressure in a reverse process by which the LiH was formed. Consequently, by increasing the temperature of the LiH sample, a higher equilibrium pressure is reached until the sample becomes depleted at that temperature and the pressure decreases. The sample decomposed very quickly above 688°C, the melting point of the two phase equilibrium pressure system, so for periods of extended data collection, the temperature was kept well below the melting temperature.

414

From the experimental results it becomes clear that the ion current collected depends not only on the temperature of the sample but also on the rate at which the temperature is changed. Figure 3 shows a typical DC ion current response to a rapid change in temperature. The current response to the rise in temperature is practically instantaneous.

For example, when the temperature of the LiH was changed by 56°C from a temperature of 587°C to a temperature of 643°C in ~ 60 s, an increase in H^- current of 17 μA was measured. The ratio of ion to electron currents, I_{H^-}/I_{e^-}, usually varied between 1/10 and 1/4 but occasionally would reach 1/100 when the rate of change of the temperature became small or static resulting in small amounts of H^- being produced. A static temperature usually produced an H^- current with a slow decay, most likely due to the depletion of the hydrogen from the sample. A test of the effectiveness of the electron trap was made with an empty sample holder using a red hot filament. No accelerated electrons were passed through the trap to the Faraday plate. The 1.2-cm-diam aperture at the entrance of the electron trap also served as the extractor electrode. The extractor was operated either as an open aperture or with a high transparency mesh across the opening. The maximum DC ion current density measured was 18 μA/cm^2 This result occurred during a moderate, positive rate of temperature change of about 1 °C/s.
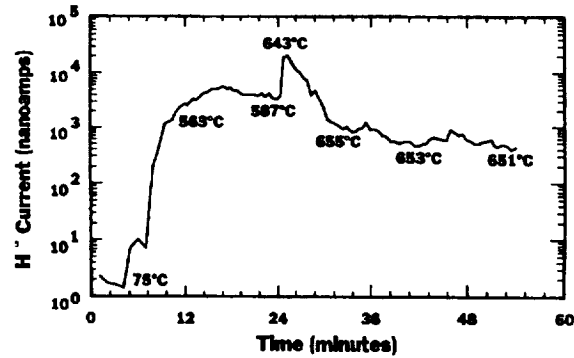

Figure 3. H^- current on Faraday plate vs time.

The results of placing an intermediate electrode between the sample and the anode (extractor) showed that it strongly affected the electron and ion currents. When a fine mesh was used as the intermediate electrode, a potential of ~ + 4 V with respect to the sample was used to maximize the H^- current. When no mesh was present on the aperture of the extractor electrode, ~ + 10 V was required on the intermediate grid electrode to maximize the electron and ion currents. When a negative potential with respect to the sample was placed on the grid or extractor electrodes, both the electron and H^- could be quenched. The variation of H^- current with extraction potential showed that the current saturated usually around 4 kV. For most of the work described here, a potential of 6 kV was used to ensure the measured currents were the maximum available.

As the temperature of the sample increased, the pressure in the vacuum system increased accordingly. In addition to H^- and electrons being emitted, hydrogen gas is released so the vacuum system must remove this unwanted gas load. Starting at a base pressure of 3 x 10^{-6} Torr, the pressure typically rose to 8 x 10^{-6} Torr during the run. Knowing the pumping speed of the vacuum pump and the conductance of the system, a gas load of approximately 4 x 10^{-2} Torr ℓ/s = 3 scc/m was estimated to be produced.

415

MASS ANALYSIS

To conclusively determine the masses of the possible ion species coming off the hydride sample, a Wein filter was constructed following the approach of Selinger[10]. Under a balanced condition, a specific ion velocity will pass straight through the analyzer with the electric field force counterbalancing the Lorentz force. Since all ions are accelerated through the same potential difference, V_0, the velocity of a given ion will depend directly on its mass. Figure 4 shows a schematic drawing of the filter. Those ions with velocity v_0 pass through the filter and a 3 mm aperture, en route to a cylindrical Faraday cup 7 cm in length. Those ions which do not possess the proper velocity are deflected as shown by the dashed line.
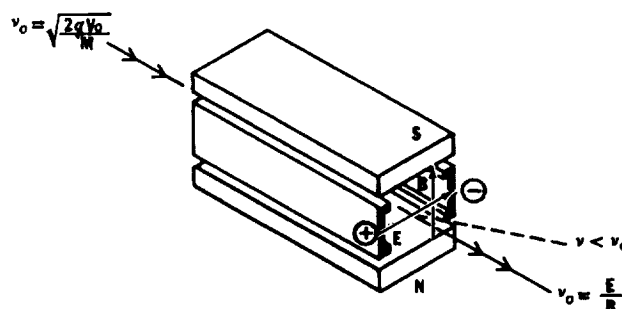
Two aluminum plates separated by 2.5 cm and machined with edge shims to reduce the fringing field were used as the electric field plates, marked + and - in Figure 4. The uniform magnetic field was provided by permanent ceramic magnets in a steel yoke with soft iron pole faces.

Figure 4. Schematic drawing of the velocity filter.

The magnetic field was mapped on four parallel axes within 1 cm of the principal axis. Figure 5 shows the strength and uniformity of the resulting field. At each end a copper plate with two sheets of mu-metal was used to terminate the E and B fields. It was found that the addition of a copper cylinder lined with mu-metal greatly reduced the B field at each end. The effective magnetic length of the mass filter was taken to be $L_m = 12$ cm. Upon exiting the filter, the ions drifted 9 cm to the aperture at the entrance of the Faraday cup. The overall resolution of the apparatus was calculated to be 10% ($\Delta m/m = 0.1$).

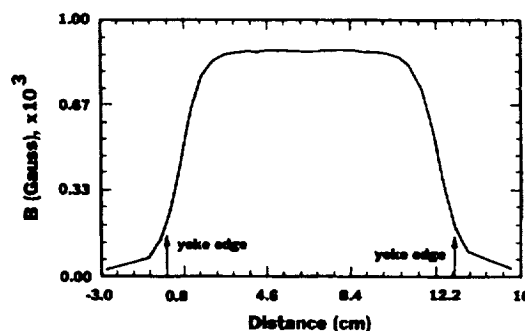**Figure 5. Mapping of magnetic field along E X B axis.**

The optics used to form a beam prior to entering the mass analyzer were designed using a cylindrical Pierce geometry[11]. This design is intended to give the ions parallel flow between the cathode and anode. Following the anode, the optics produced an ion beam with a focal length of $f = -3$ cm. The lithium hydride sample of was heated in the container below the Pierce electrode. The anode surface was formed from copper with a radius of 2.5 cm. For an aperture of $r_0 = 2$ mm, the half angle divergence of the beam is 3.8°. The entire anode-cathode assembly is housed in a steel yoke so that small Nd Fe magnets could be used to remove electrons generated in the emission

416

process if desired. The mapping of the magnetic field in this region produced a value of $\int Bdl = 450$ G cm. with an average value of $B = 340$ G which is sufficient to remove 6 keV electrons. Due to the small temperature gradient applied to the sample during the mass analysis, small ion currents were produced, so an einzel lens was constructed to increase the intensity of the ions prior to entering the mass analyzer. This reduced the drift distance following the analyzer to 5 cm but did not significantly affect the resolution of the apparatus.

A series of potential difference readings across the electric field plates in the analyzer were made at each of four accelerating voltages, V_0. Each series of potential differences placed across the analyzer plates required so much time to complete that the rate of temperature change of the sample had to be kept at a small value to keep the sample from releasing the stored hydrogen too quickly. Consequently, the amount of H^- produced at the source, as analyzed by the Wein filter and passed through the 3-mm-diam aperture to the Faraday cup, was small for the mass analysis measurements. As noted earlier, a positive rate of temperature change is required to produce a significant ion current. Therefore, it is not surprising that under the conditions of nearly static temperatures, the amount of collected ion current would be much reduced from the earlier experiments where larger rates of temperature change were maintained. A typical voltage sweep is shown in Figure 6.

Because the power supply providing the accelerating voltage was not regulated and tended to slowly drift, a slightly larger FWHM was produced on the peak than expected. When using the Wein filter for the mass analysis no electron trap was required since the electron experiences a much larger magnetic force compared to the ions and hence is promptly removed upon entering the crossed fields of the analyzer. The power supplies which provided the voltages for the crossed field analyzer handled the increased electron current loading without a significant drop in potential. Using the condition for the balance of forces in the analyzer and conservation of energy gives
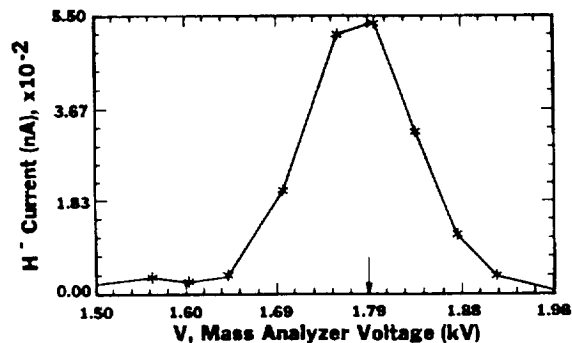

Figure 6. H$^-$ current collected by Faraday cup as electric field is changed in E X B analyzer. Voltage uncertainties are ± 50 V.

$$V^2 = 2B^2 d^2 \frac{q}{m} V_0 \tag{1}$$

where V is the potential difference across the E-field plates, V_0 the accelerating potential of ion beam, B the magnetic field in analyzer, d the separation of E-field plates (2.54 cm), q/m the ionic charge to mass ratio, and where $2B^2 d^2 = 9.99 \times 10^{-6}$ T^{2}m^2.

417

A plot of V^2 vs V_0 in Figure 7 shows the slope of the experimental data is best matched by ions with a charge to mass ratio, corresponding to H^-. This plot is produced from the analyzer peak voltages which result from the four voltage sweeps made on the Wein analyzer. The peak voltage was estimated as a centroid in the plane of each curve. A more accurate least square fitting of each sweep could generate a slightly different peak voltage but that would not alter the clear match to the data by ions with $M = 1$. The possibility of negative ions with mass $M = 2$ being produced and passed through the Wein analyzer would require a charge of $q = -2$ in order to maintain the same charge to mass ration in Eq. (1). As part of the series of voltage sweeps on the analyzer, the possible presence of a $M = 2$ peak located several hundred volts away from the $M = 1$ peak was investigated. As expected, since H^{--} is not a stable ion, no peak was observed. Therefore, we conclude that H^- is the ion species observed as LiH is heated.
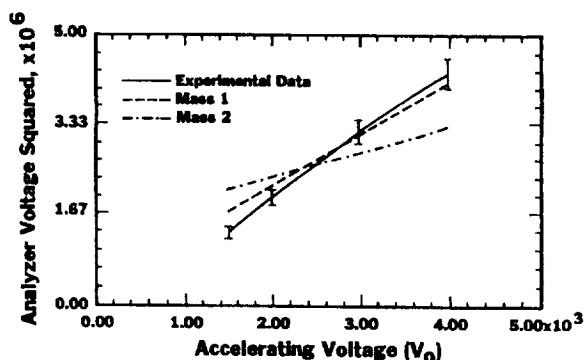

Figure 7. Graph of experimental data in Eq. (1). Accelerating voltage uncertainties are ± 75 V.

DISCUSSION AND FUTURE WORK

With the identity of the emitted negative ions confirmed, the logical next step is to understand the mechanism involved with the production of the H^-. It has been known for some time that above 250°C, LiH decomposes in a vacuum. At the temperatures used in this work, the lithium metal evaporates since it has an equilibrium pressure much larger than the surrounding vacuum.[12] Normally, when LiH is heated, the hydrogen is liberated to form molecular hydrogen. However, in this experiment, the H^- may actually be emitted directly from the LiH surface since the sample is connected to a power supply which delivers sufficient negative charge to neutralize the Li^+ prior to the Li being evaporated. This is consistent with low-energy electron diffraction (LEED) measurements made by Powell et al.[13], which shows that as hydrogen is removed, the surface unit mesh increases relative to the bulk unit mesh and the stoichiometry of the LiH system changes so the surface becomes enriched in lithium.

An alternate view of the H^- emission involves a surface process in which the outgoing H atoms thermally liberated from the sample become ions by the tunneling of electrons from the conduction hand of the metal to the affinity level of the H atom. In this model the frequency of tunneling depends strongly on the density of surface electrons and the work function of the surface. Several experiments[14-16] have shown that low-energy (~ 1 eV) H atoms may reach an ionization probability greater than 0.1 when the work function of the converter material is less than 1.5 eV. In the case of LiH the value of the work function does not appear to be available in the scientific literature. However, on the basis of a model developed by Yamamoto et al[17] used to calculate the work function of a binary compound

418

involving gas absorption on a metal surface, the work function for the hydride is likely not much less than that of Li (2.4 eV). Consequently, it is not clear that the resonant charge transfer model should be used to interpret the H^- formation near the LiH surface.

Whatever the precise nature of the production mechanism, it is clear that this technique has the potential to achieve three important goals for future applications requiring sources of negative hydrogen ions. First, by using other saline hydrides such as NaH, which has a minimum equilibrium hydrogen pressure of 5800 mm at 500°C, the corresponding negative ion emission should be significantly increased. Increasing the temperature to 550°C raises the equilibrium pressure almost a factor of 3 beyond the 5800 mm value.[6] Consequently, the prospect of developing a more elementary DC negative hydrogen ion source with an output current density at least comparable to those source already developed would appear to have possible commercial merits.

Secondly, the transport of the negative ions to the region in space where they are neutralized and eventually used requires a low value of transverse emittance. The present generation of H^- sources produce ion beams which possess emittance values much lower than those from a decade earlier. However the techniques described here involving a sample at ~ 700°C should result in an even smaller value. To operate at a temperature of only 500°C in the case of NaH would produce an even lower value of emittance for the ion/neutral beam being transported.

Thirdly, the lifetime of a saline hydride ion source would not be limited by the lifetime of a filament as is true for many present day sources. The hydride could be continuously fed into the heater or hydrogen gas could be used to resupply the metal hydride to keep it from being depleted of hydrogen. Either method should produce a long lifetime for the source. These are matters to be decided upon further technical development.

Ion sources usually require several years of study and advanced development before they become optimized. The technique described here is no exception. The work presented here notes the possibility of creating an advanced negative hydrogen ion source with many desirable features which may be of benefit as we face the energy needs of the next century.

References

1. M.K. Hubbert, <u>Resources and Man</u>, (W.H. Freeman, 1969).

2. H.P. Eubank et al., Phys. Rev. Lett. <u>43</u>, 270 (1979).

3. D.J. Rej, I. Henins, R.J. Fonek, and Y.J. Kim, Rev. Sci. Instrum. <u>63</u>, 4934(1992).

4. K.W. Ehlers, K.N. Leung, R.V. Pyle and W.B. Kunkel, <u>Proceedings of the Fourth International Symposium on the Production and Neutralization of Negative Ions</u>, AIP NO. 158, Brookhaven, 1986, p. 282.

5. G.G. Libowitz, in <u>Critical Materials Problems in Energy Production</u>, (Academic Press, New York, 1976).

6. W.M. Mueller, J.P. Blackledge and G.G. Libowitz, <u>Metal Hydrides</u>, (Academic Press, New York, 1986).

7. R.S. Calder, W. Cochran, D. Griffiths, and R.D. Lowde, J. Phys. Chem. Solids <u>23</u>, 621 (1962).

8. F.E. Pretzel, G.N. Rupert, C.L. Mader, E.K. Storm, G.V. Gritton, and C.C. Rushing, J. Phys. Chem. Solids <u>16</u>, 10 (1960).

9. R.L. Palmer, <u>Proceedings of the Third International Symposium and Neutralization of Negative Ions</u>, AIP NO. 111, Brookhaven, 1983, p. 281.

10. R.L. Seliger, J. Appl. Phys. <u>43</u>, 2352 (1972).

11. A.T. Forrester, <u>Large Ion Beams</u>, (Wiley, New York, 1976).

12. C.E. Holcombe, G.L. Powell, and R.E. Clausing, Surf. Sci. <u>30</u>, 561, (1972).

13. G.L. Powell, G.E. McGuire, D.S. Easton, and R.E. Clausing, Surf. Sci. <u>46</u>, 345 (1974).

14. A. Pargellis and M. Seidl, Phys. Rev. B<u>25</u>, 4356 (1982).

15. J.N.M. Van Wernnik, J.J.C. Geerlings, and J. Los, Surf. Sci. <u>131</u>, (1983).

16. J.P. Gauyacq and J.J. C. Geerlings, Surf. Sci. <u>182</u>, 245 (1987).

17. S. Yamamoto, K. Susa, U. Kawage, and H. Okamo, Jpn. J. Appl. Phys. Suppl. <u>2</u>, 209 (1974).