SAND2017-10151R

Built on LDRD Laboratory Directed Research and Development

.

SAND20XX-XXXXR
LDRD PROJECT NUMBER: 201112
LDRD PROJECT TITLE: A Hydrogen and He Isotope Nanoprobe
PROJECT TEAM MEMBERS: Barney L. Doyle and Stuart B. Van Deusen, both 01861

ABSTRACT:

Materials that incorporate hydrogen and helium isotopes are of great interest at Sandia and throughout the NNSA and DOE. The Ion Beam Lab at SNL-NM has invented techniques using micron to mm-size MeV ion beams to recoil these light isotopes (Elastic Recoil Detection or ERD) that can very accurately make such measurements. However, there are many measurements that would benefit NW and DOE that require much better resolution, such as the distribution of H isotopes (and ^{3}He) in individual grains of materials relevant to TPBARs, H and He-embrittlement of weapon components important to Tritium Sustainment Programs, issues with GTSs, batteries… Higher resolution would also benefit the field of materials science in general. To address these and many other issues, nm-scale lateral resolution is required. This LDRD demonstrated that neutral H atoms could be recoiled through a thin film by 70 keV electrons and detected with a Channeltron electron multiplier (CEM). The electrons were steered away from the CEM by strong permanent magnets. This proved the feasibility that the high energy electrons from a transmission-electron-microscope-TEM can potentially be used to recoil and subsequently detect (e-ERD), quantify and map the concentration of H and He isotopes with nm resolution. This discovery could lead to a TEM-based H/He-isotope nanoprobe with 1000x higher resolution than currently available!

INTRODUCTION:

Issues involving hydrogen-isotopes are of great interest at Sandia and other NW labs. This LDRD explored using the nm resolution capability of a TEM to quantitatively measure and map H and He isotopes. While these elements are the most abundant in the universe, they are also the most difficult to quantitatively measure in solids. The Ion Beam Lab (IBL) at SNL-NM has invented techniques using micron to mm-size MeV ion beams to recoil these light isotopes (Elastic Recoil Detection or ERD) that can very accurately make such measurements. However, there are many measurements that would benefit NW that require much better resolution, such as the distribution of H isotopes (and ^{3}He) in individual grains of materials relevant to TPBARs, H and He-embrittlement of weapon components important to Tritium Sustainment Programs, issues with GTSs, batteries… Higher resolution would also benefit the field of materials science in general. To address these and many other issues, nm-scale lateral resolution is required.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Over the past three years, radical proposals have been made that >100 keV electrons from a TEM could be used to recoil and subsequently detect (e-ERD) and quantify the concentration of H and He isotopes with nm resolution. This would lead to a TEM-based H/He-isotope nanoprobe with 1000x higher resolution than currently available! While LDRD proposals have been submitted starting in 2013, only this current 2017 small Exploratory Express LDRD has been funded. The ultimate goal of this project was then to prove the feasibility of the e-ERD technique and provide recommendations on how to realize a Hydrogen and He Isotope Nanoprobe. **The project was successful**.

In the remainder of this INTRODUCTION section, the physics of the electron-nucleus interaction is reviewed and the equations for calculating both the energy and cross section (scattering probability) of H atoms by energetic electrons are given. These equations were central in designing the prototype system that was tested in the QASPR-III endstation in the target room of the Tandem accelerator in the IBL. These equations were also useful for predicting the performance of the prototype e-ERD system.

The DETAILED DESCRIPTION OF EXPERIMENT/METHOD section of this report the design, theory of operation and anticipated performance of the prototype e-ERD system is described.

In the RESULTS section, the results of the first tests of the e-ERD concept involving successful measurements of H in Mylar are given.

The DISCUSSION section analyzes the results reported in the previous section and makes comparisons to the theory giving in the Introduction.

A summary, recommendations for improvements and a path forward for realizing an e-ERD system on a Sandia TEM are given in the ANTICIPATED OUTCOMES AND IMPACTS section.

Theory of Electron-Nucleus Collisions

The physics of the electron-nucleus collision is fairly straightforward and has been studied for over 100 years [1]. The development that follows is therefore hardly original but is included for completeness. Ultimately what is needed are calculations of the recoil energies and cross sections made by energetic electrons, in addition to the energy loss suffered by these recoils in the material from which they are recoiled. This is precisely the same information that is needed to calculate electron radiation damage effects in materials, and James Corbett wrote an excellent book in 1966 titled "Electron Radiation Damage in Semiconductors and Metals" [2] which was part series of books edited by F. Seitz and D. Turnbull [3]. The mathematical development in Corbett's book is closely followed here due to the similarities of both problems. Figure 1 shows a scattering diagram of an energetic electron recoiling a nucleus in both the center of mass and laboratory frames.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

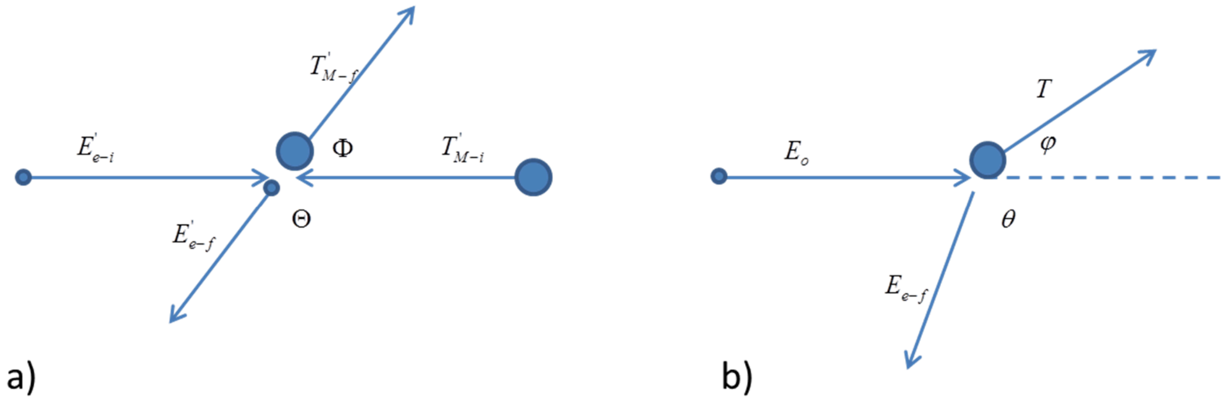

Figure 1: Scattering diagram of electron (small circles) on atomic nucleus (large circles) in a) the center of mass (com) frame and b) the laboratory (lab) frame. The incident and final energies of the electrons are E' and E in the com and lab frame respectively, and T' and T for the nucleus that is recoiled.

Kinematics

For the detailed mathematical derivation of the kinematics of the electron-nucleus interaction the reader is referred to the Corbett book [2]. However there are mistakes in some of his equations, and those will be pointed out here.

The kinetic energy of a nucleus of mass M recoiled by a relativistic electron of mass m and initial energy of E_o at an angle φ in the lab frame is given by:

$$T = \frac{2p_o^2 cos^2(\varphi)}{M} \tag{1}$$

(note: in Corbett eq. 4.22 the M in the denominator should not be squared)

p_o is the relativistic momentum of the electron. The maximum recoil energy given to the nucleus is at a recoil angle of 0^o , and this energy can be expressed as:

$$T_{max} = \frac{2p_o^2}{M} = \frac{2(\gamma m v_o)^2}{M} = 2\frac{m}{M}\gamma^2\beta^2 mc^2 \tag{2}$$

and

$$\gamma = \frac{1}{\sqrt{1-\beta^2}} = 1 + \frac{E_o}{mc^2},\ \beta = \frac{v}{c} = \sqrt{1 - \frac{1}{\gamma^2}}, \tag{3}$$

It is also useful to express the recoil energy as:

$$T = T_{max} \sin^2 \frac{\Theta}{2} \tag{4}$$

The maximum recoil energy of a proton recoiled by an electron is plotted in Figure 2.

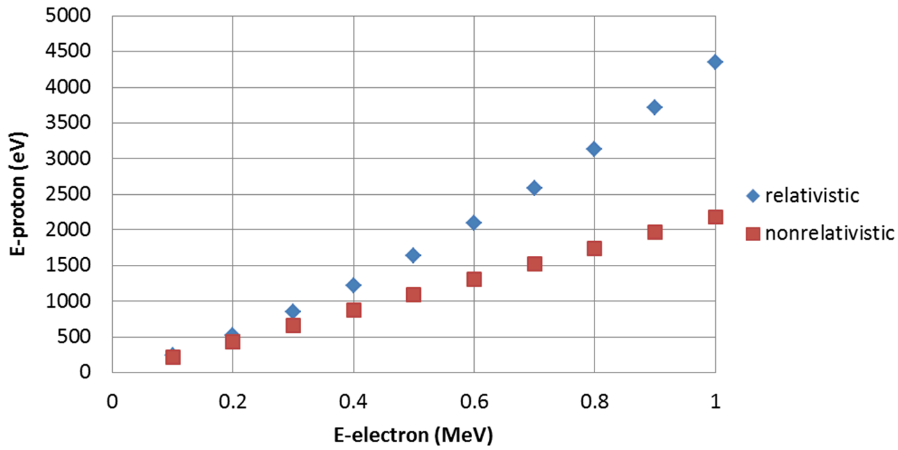

Figure 2: Maximum recoil energy of protons by electrons with initial kinetic energies from 0.1 to 1.0 MeV. The relativistic calculation becomes very important at energies greater than ~1/2 the .511 MeV rest mass of the electron.

70 keV electrons were used to recoil protons in the experiment described below, and in this case the maximum recoil energy is 164 eV.

Recoil Cross Sections

Rutherford's famous nonrelativistic differential cross section equation for Coulomb scattering which was derived in reference [1] for electrons scattering from a nucleus of atomic number Z is:

$$d\sigma_R = \left(\frac{Ze^2}{2mv^2}\right)^2 \frac{1}{\sin^4 \frac{\Theta}{2}} d\Omega \quad , \tag{5}$$

where Ω is the solid angle for an annular cone with a width $d\Omega$ about Ω:

$$d\Omega = 2\pi \sin\Theta d\Theta = 4\pi \sin\frac{\Theta}{2} \cos\Theta d\Theta \tag{6}$$

Built on LDRD Laboratory Directed Research and Development

The Rutherford cross section then becomes:

$$d\sigma_R = \pi\left(\frac{Ze^2}{mv^2}\right)^2 \frac{\cos\frac{\Theta}{2}}{\sin^3\frac{\Theta}{2}} d\Theta \quad . \tag{7}$$

Darwin later modified Rutherford's cross section for the case of relativistic incident particles deriving the Dirac-Rutherford equation:

$$d\sigma_{DR} = \pi\left(\frac{Ze^2}{mc^2}\right)^2 \left(\frac{1}{\beta^4\gamma^2}\right) \frac{\cos\frac{\Theta}{2}}{\sin^3\frac{\Theta}{2}} d\Theta \quad . \tag{8}$$

(note: in Corbett eq. 4.31 had the Lorentz factor as γ^4 and it should have been γ^2)

Further improvements were made to this equation by treating the scattering quantum mechanically. For the recoiling of light nuclei, i.e. Z and M are small (which is the case we are interested in here), McKinley and Feshbach derived an approximate quantum mechanical recoil cross section:

$$d\sigma_{McF-DR} = \pi\left[1-\beta^2\sin^2\frac{\Theta}{2} + Z\left(\frac{e^2}{hc}\right)\beta\pi\sin\frac{\Theta}{2}\left(1-\sin\frac{\Theta}{2}\right)\right]\left(\frac{Ze^2}{mc^2}\right)^2 \left(\frac{1}{\beta^4\gamma^2}\right) \frac{\cos\frac{\Theta}{2}}{\sin^3\frac{\Theta}{2}} d\Theta \quad . \tag{9}$$

For our case, and Corbett's, expressing the recoil cross section differential in recoil energy is most convenient. Differentiating equation 4 above yields:

$$dT = T_{\max}\sin\frac{\Theta}{2}\cos\frac{\Theta}{2}d\Theta \quad , \tag{10}$$

and substituting equations 10 and 4 into equation 8 provides the Dirac-Rutherford equation differential in recoil energy:

$$d\sigma_{DR} = \pi\left(\frac{Ze^2}{mc^2}\right)^2 \left(\frac{1}{\beta^4\gamma^2}\right) T_{\max}\frac{dT}{T^2} \quad . \tag{11}$$

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

To calculate the cross section for recoiling a nucleus in an energy range between T_{min} and T_{max} one simply integrates equation 11, evaluating at these limits:

$$\sigma_{DR}(T_{\min-\max}) = \pi\left(\frac{Ze^2}{mc^2}\right)^2\left(\frac{1}{\beta^4\gamma^2}\right)T_{\max}\int_{T_{\min}}^{T_{\max}}\frac{dT}{T^2} = \pi\left(\frac{Ze^2}{mc^2}\right)^2\left(\frac{1}{\beta^4\gamma^2}\right)\left(\frac{T_{\max}}{T_{\min}}-1\right). \quad (12)$$

With a little more effort, the same can be done for the McKinley-Feshback equation:

$$\sigma_{McF} = \pi\left(\frac{Ze^2}{mc^2}\right)^2\frac{1}{\beta^4\gamma^2}\left[\left(\frac{T_{max}}{T_{min}}-1\right)-\beta^2\ln\left(\frac{T_{max}}{T_{min}}\right)+\pi\alpha\beta\left\{2\left[\left(\frac{T_{max}}{T_{min}}\right)^{1/2}-1\right]-\ln\left(\frac{T_{max}}{T_{min}}\right)\right\}\right]. \quad (13)$$

A comparison of these two recoil cross sections for electrons recoiling protons is given in Figure 3 below.

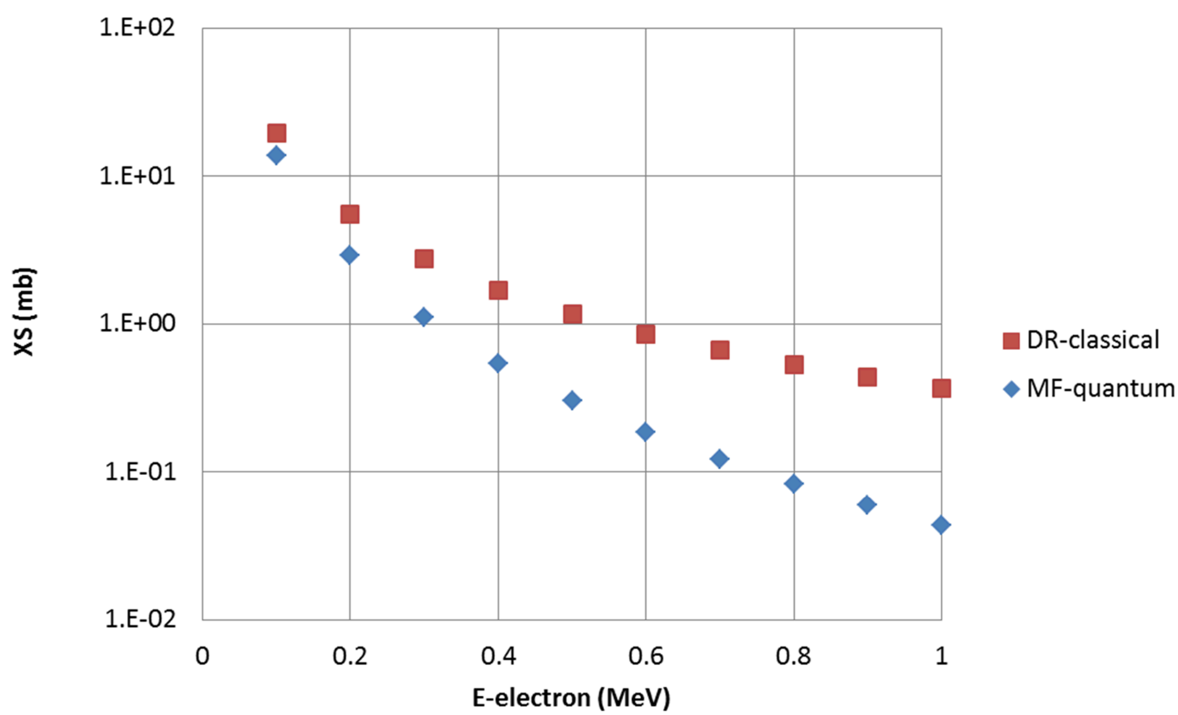

Figure 3: Classical (Darwin-Rutherford) and Quantum Mechanical (McKinley-Feshbach) calculations of electron cross sections for electrons from 0.1 to 1.0 MeV to recoil protons to between 99-100% of the maximum recoil energies, i.e. T_{min}=0.99T_{max}. This corresponds to recoiling the protons into a cone angle of 5.7°.

It obviously becomes important to use the quantum mechanical calculation for the higher energy electrons. 70 keV electrons were used to recoil protons in this experiment, and in this case the cross section using the Mckinley-Feshbach equation is 29 mb or 2.9e-26 cm^2.

DETAILED DESCRIPTION OF EXPERIMENT/METHOD:

The electron elastic recoil detector

In this section, the design and modeling of the first prototype e-ERD detector is described. This system had to be quite small in order to fit on the XY stage of the QASPR-III endstation in the Tandem target room of the IBL. This site was selected for these initial tests instead of modifying a TEM for several reasons:

1. A 100 keV electron gun was already installed on this endstation for radiation effects testing
2. There was already space available in the endstation where the detector could be installed
3. Only minor vacuum feedthrough modifications were required to provide the HV bias to the detector and signal cables.

It was decided that these initial system tests would be performed using 70 keV electrons and with 0.5 micron thick Mylar targets because they were transparent to the electrons and contained a significant amount of H, $C_{10}H_8O_4$.

e-ERD Detector Design

There were several requirements that the e-ERD detector had to fulfill: it had to:

1. be small, <5" to fit between the QASPR-III stage and the focal point of the front viewing microscope
2. support the Mylar target that contained the H to be recoiled
3. steer the electrons away from the final detector
4. this final detector needed to detect low energy (<164 eV) H, most likely neutral atoms

To satisfy these conditions, the components were fabricated into three chambers in a small "Bud" box as shown in Figure 4. A hole was drilled and a 3mm TEM target holder was attached to the front of the box. The transmitted electrons and recoiled H atoms then traveled 24mm in Chamber 1 where they then passed through a 3mm diameter aperture and into the 18mm region of Chamber 2 that had a 2kG magnetic field produced by two 18x18mm square 1kG permanent magnets. The recoiled H atoms then passed through another aperture 6mm in diameter in to Chamber 3, but the electrons were to be prevented from passing through this final aperture due to the strong magnetic field. The final detector that was selected was a commercial Channeltron electron multiplier [5]. The trajectories of the electrons (red), neutral H atoms (black) and positive H^+ ions (blue) were modeled using the SIMION program [6]. These calculations show that the magnetic field was more than adequate to steer the electrons away from the second aperture preventing them from being detected by the Channeltron.

While data for the efficiency of Channeltrons to low energy H ions or atoms was not found, there were papers on the efficiency of Channelplates [7,8], which are quite similar. Data from Reference 7 is plotted in Figure 5. For H atoms in the 100 eV energy range the efficiency is seen to be ~ 10%. As a side note, one of the authors of reference 8, D. H. Crandall, was the Chief Scientist at NNSA who approved the construction of the new IBL.

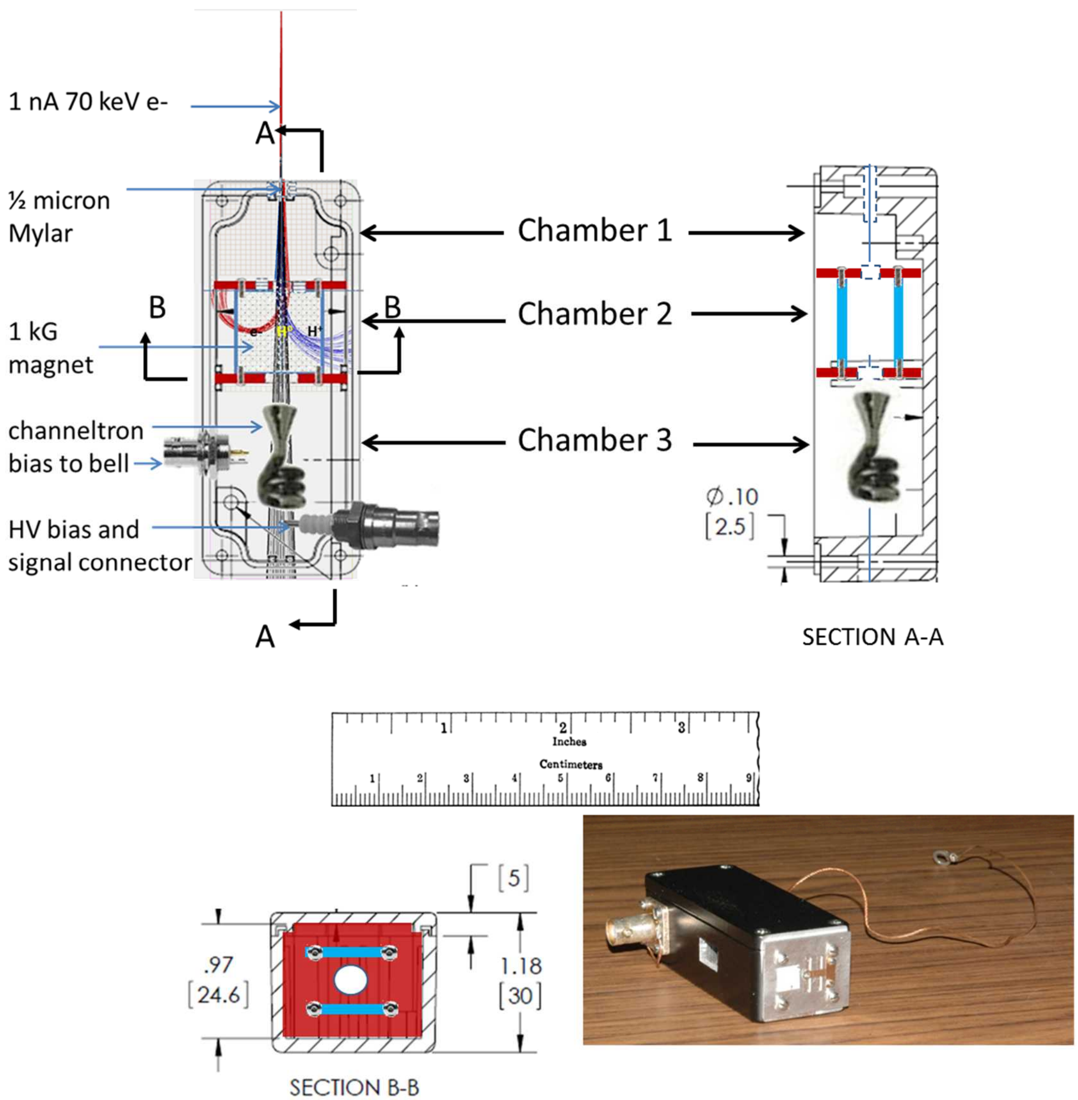

Figure 4: Design drawings of the e-ERD target holder and detector. The various components of the detector are indicated in the upper right, and the trajectories of the electrons (red), neutral H atoms (black) and positive H^+ ions (blue) were modeled using the SIMION program.

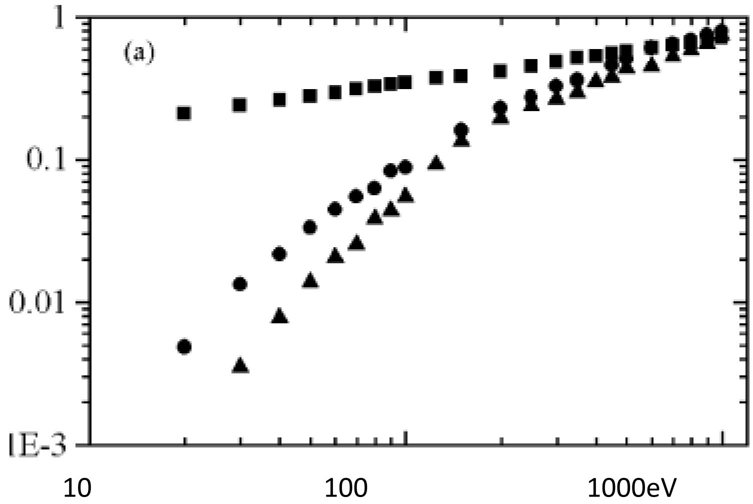

Figure 5: Detection efficiency of a Channelplate for low energy H^- (squares), H^o (circles), and H^+. The energy of the recoiled H^o in this experiment is 164eV, and the efficiency is therefore 0.1 or 10%. (After Peko and Stephen, Ref. 7)

Electronics and Signal Processing

Standard nuclear-based electronics was used to process the signals made when H^o atoms struck the Channeltron. The bell was biased at -100V and the signal end biased at +2000V through the HV preamplifier. Those signals were further amplified with a spectroscopic amplifier set for 10µs shaping time to reduce ringing. The bipolar output pulses of the spectroscopic amplifier were then sent to a multichannel analyzer. The electronics are shown in Figure 6.

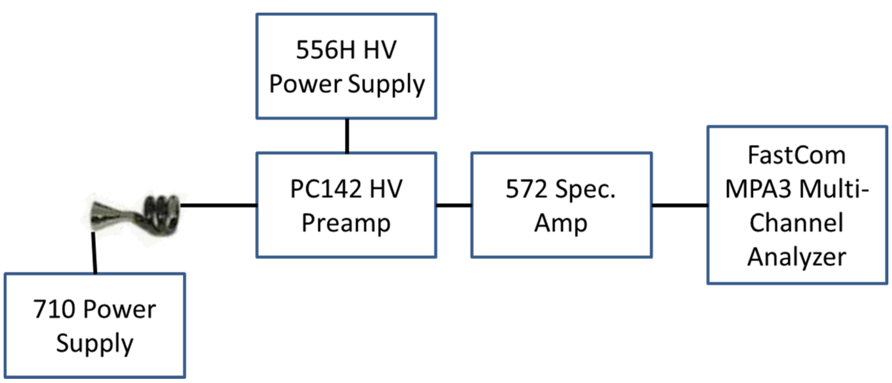

Figure 6: Electronics used to bias and process the Channeltron signals. All of the power supplies and amplifiers were made by Ortec.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Targets

Two targets were selected for the e-ERD tests, one that contained H (Mylar) and the other that didn't (Au).

Mylar Target

Both targets were alternately mounted on the e-ERD detector and attached to the QASPR-III XY stage, and positioned to be exposed to the 70 keV electron beam from the e-gun.   A micrograph obtained by an "in-line" microscope is shown in Fig. 7 of the Mylar sample with a superposition of the electron beam added. It is clear that the electron beam has been focused to be considerably smaller than the 3mm diameter targets.

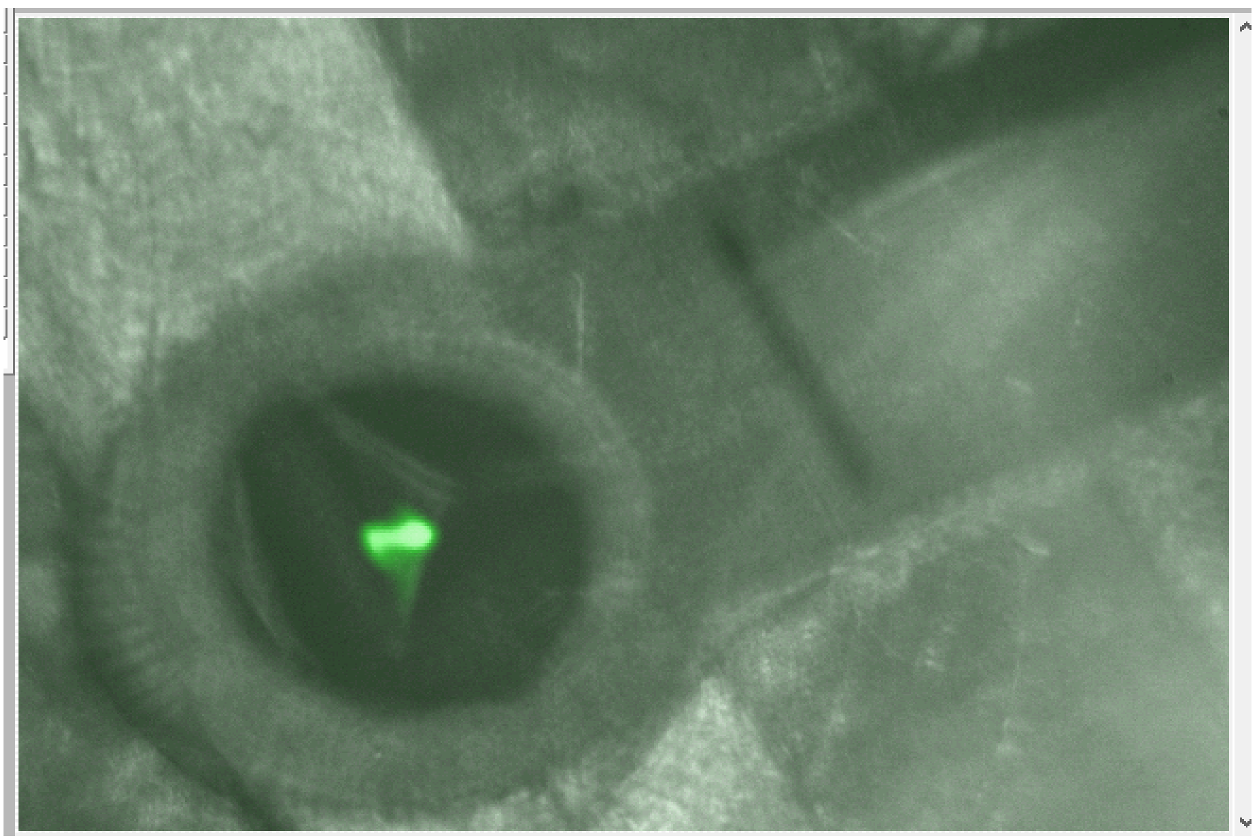

Figure 7: Micrograph of the Mylar sample with a superposition of the electron beam.

Built on
LDRD
Laboratory Directed Research and Development

The Mylar was 0.5 microns thick and had a high H concentration of 3.51E+22/cm3. A plot of the energy of the recoiled H atoms was calculated using the ZBL-LSS stopping power theory [9] is shown in Fig. 8.

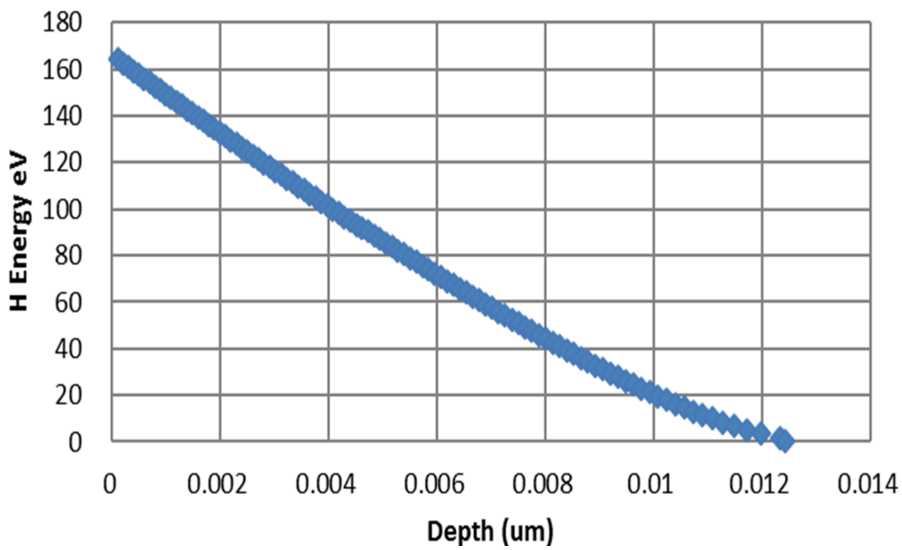

Figure 8: Calculation of the energy of the H recoiled by 70 keV electrons as they exit the backside of the Mylar foil.

The highest energy H is recoiled from the back of the Mylar into the e-ERD detector, while the lowest energy H is recoiled 0.012 microns from the backside.

It is useful to estimate here the signal strength of the recoiled H. The equation for the Yield per second of H recoiled into the e-ERD detector is:

$$Y \approx \phi C_H t \sigma \varepsilon \tag{14}$$

Where, using the McKinley-Feshbach recoil cross section for a μA of 70 keV electrons:

$$\phi = 1\mu A = 6.25x10^{12} e/s$$
$$C_H = 3.5x10^{22} H/cm^3$$
$$t = 1.2x10^{-6} cm$$
$$\sigma = 2.9x10^{-26} cm^2$$
$$\varepsilon = 0.1$$

This gives a detected H rate of 776 H/µC or 0.776 H/nC, which should be easily detectable.

Au Target

The Au target was mounted on a TEM grid and had a thickness of 0.08 micron. There would be only a monolayer of H on the surface of this sample, and any signals would be considered background count rates for the e-ERD measurement. Background count rate measurements were also to be performed with no target inserted into the holder.

RESULTS:

A photograph of the e-gun and QASPR-III endstation is shown in Fig. 9 together with a schematic diagram of the entire system. Experiments where 70 keV electrons were exposed to the various targets measuring the signals from the Channeltron were carried out during the 2nd and 3rd quarter of FY17. As indicated above only minor modifications such as adding SHV BNC feedthroughs and hardware to attach the e-ERD system were required of the QASPR-III endstation. Nevertheless, it was very important that these experiments did not interfere with the important work of the QASPR program, and special care was taken during the scheduling, equipment installation and e-gun operation processes to avoid this interference.

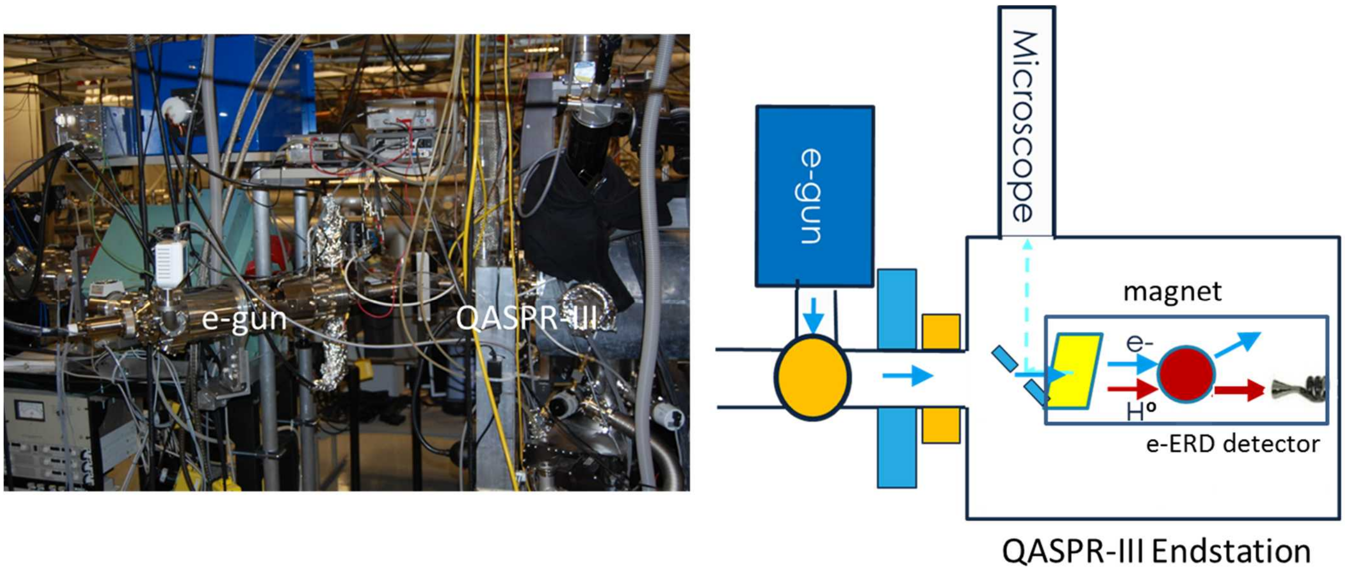

Figure 9: Photograph and schematic of the QASPR-III endstation system modified to perform the e-ERD experiments.

The basic idea behind the experiments was to measure the 70 keV electron beam current and determine the corresponding count rate of signals coming from the Channeltron for both targets that contained H (Mylar) and those that did not (Au and "open"). This count rate adjusted for background is then compared to that calculated using Eq. 14.

The electron beam current was measured in a faraday cup upstream from the QASPR-III endstation. The e-beam was pulsed so as to minimize the chance of heating or worse, destroying the Mylar foil. The current as processed through a transimpedance amplifier (TIA) set at $1A=10^8V$ (i.e. 1V=10nA) is plotted for a 100μs pulse in Fig. 10. Because the count rate for the Mylar target was expected to be low, the initial currents were only in the μA range, but we were surprised to see an extremely high count rate, and therefore the current was reduced to be in the nA range.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

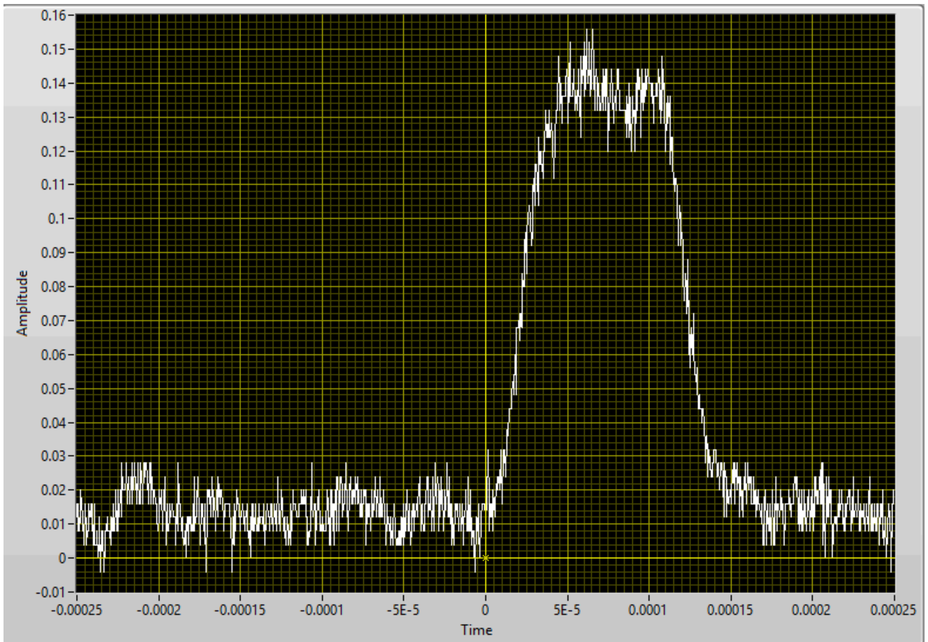

Figure 10: 100 microsecond pulse of 70 keV electrons measured in the Faraday cup upstream from the QASPR-III endstation. 0.01V amplitude indicates a current of 1 nA.

The Channeltron was biased at +1800V and the bell at -300V. The signals were then processed as shown in Fig. 6, and a representative spectrum of counts vs. channel number in the MCA data is plotted in Fig. 11.

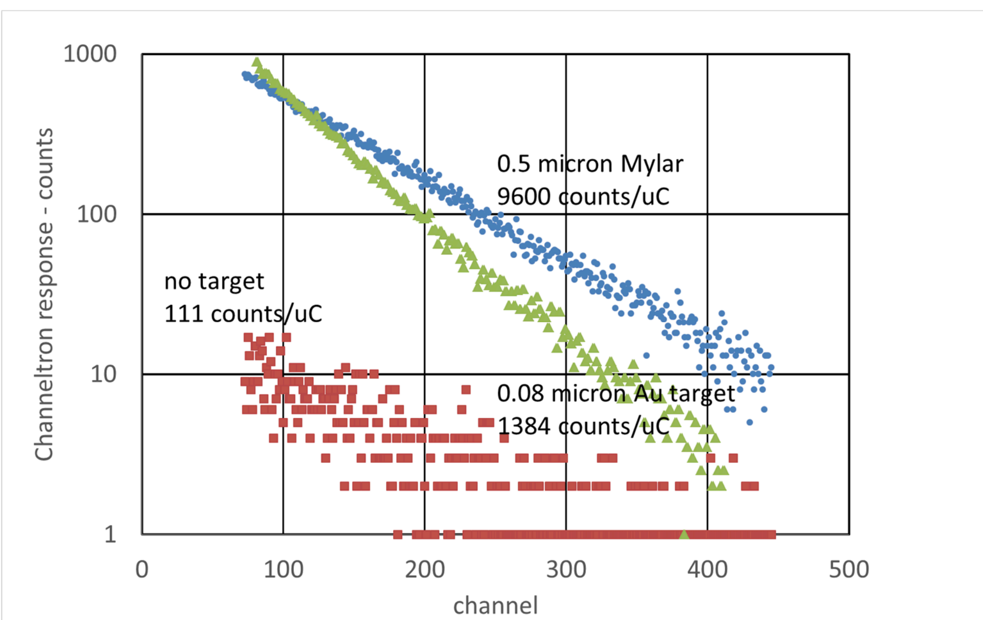

Figure 11: Amplified Channeltron signal output for 1nA of 70 keV electrons placed on 0.5 um Mylar foil (blue), a 0.08 um Au foil (green) and with no target (red) for 50 seconds.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

DISCUSSION:

From the theory presented above, the yield of H atoms recoiled from the 0.5 micron Mylar foil was expected to be .776 H/uC and nearly 0.0 for the Au foil and open target. The yields plotted in Fig. 11 are 100s to 1000s of times greater. It was therefore clear that the 1.9 kG magnetic field in Chamber 2 was not keeping electrons from entering Chamber 3, and causing counts in the Channeltron.

SIMION calculations using many more electron trajectories than before verified this problem and is plotted in Fig. 12. The trajectory of direct 70 keV e's with no scattering in the target or aperture 1 is shown in Panel A. These were the trajectories that were to have no electrons passing through aperture 2 into Chamber 3 and preclude electrons from causing backgrounds in the Channeltron. But subsequent SIMION calculations included simulated scattering from both the target and aperture 1 and showed that electrons could find trajectories that could pass through aperture 2 into the Channeltron, and this is shown in Panel B. The solution to this high background problem was found in Panel C, where a 4kG magnetic field was placed in Chamber 1 of the e-ERD system. This field steered the main beam of electrons away from aperture 1, nearly eliminating the scattering of electron by this aperture which was the main source of Channeltron backgrounds.

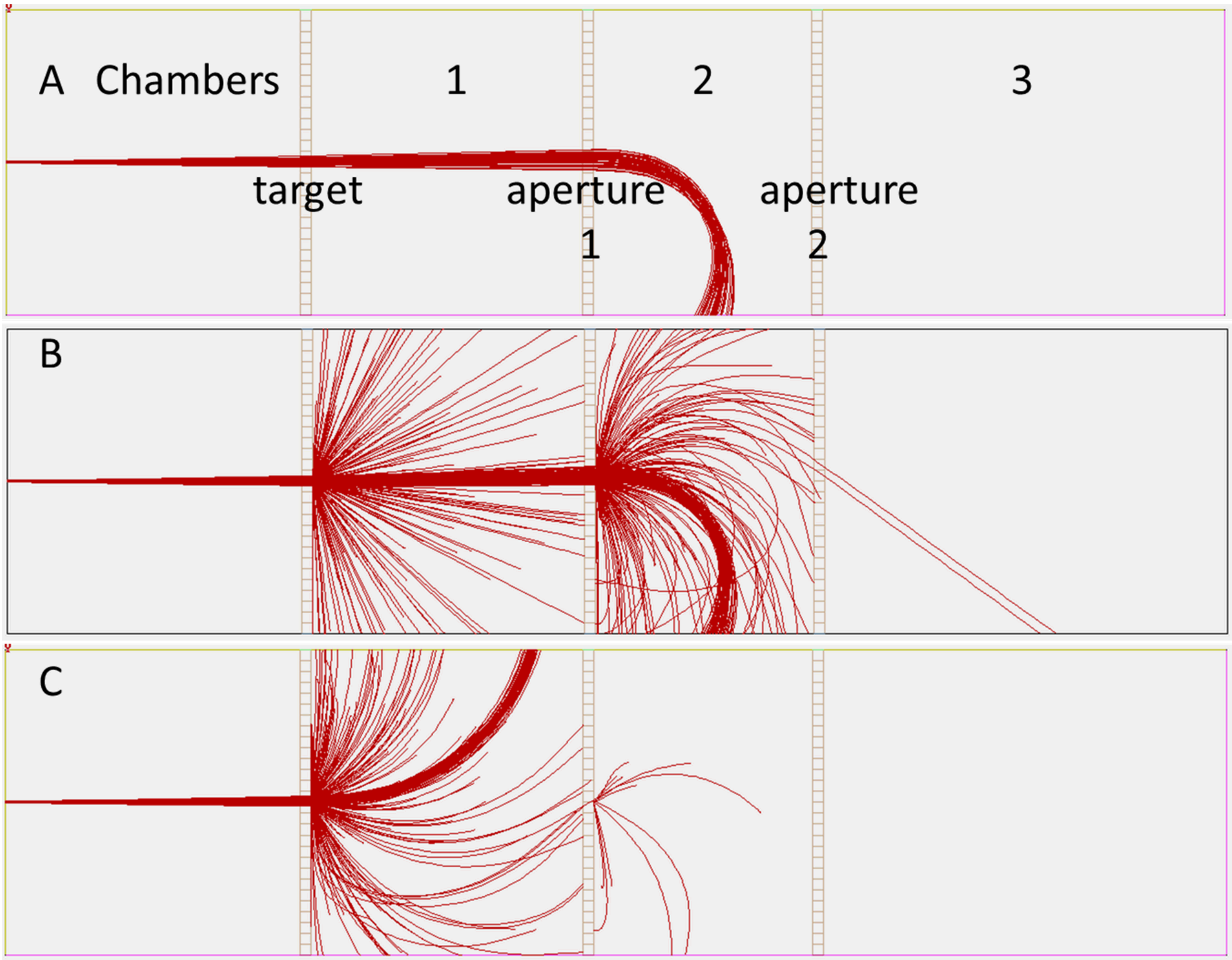

Figure 12: SIMION calculations electron trajectories for three situations of the e-ERD detector system. A: 1.9 kG magnetic field in Chamber 2 but no scattering of electrons from target or aperture 1, B: same but includes scattering at target and aperture 1, C: same 1.9 kG magnetic field in Chamber 2, but also a 4 kG field in Chamber 1 including electron scattering.

The Channeltron signal for the e-ERD detector system with magnetic fields in both Chambers 1 and 2 is plotted in Fig.13 for 70 keV electrons on the Mylar, Au and open targets. The backgrounds in this case have been eliminated and the counts for the and open target cases are just the dark noise of the Channeltron plus a few real H recoils for the Au target due to surface monolayers on the foil.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

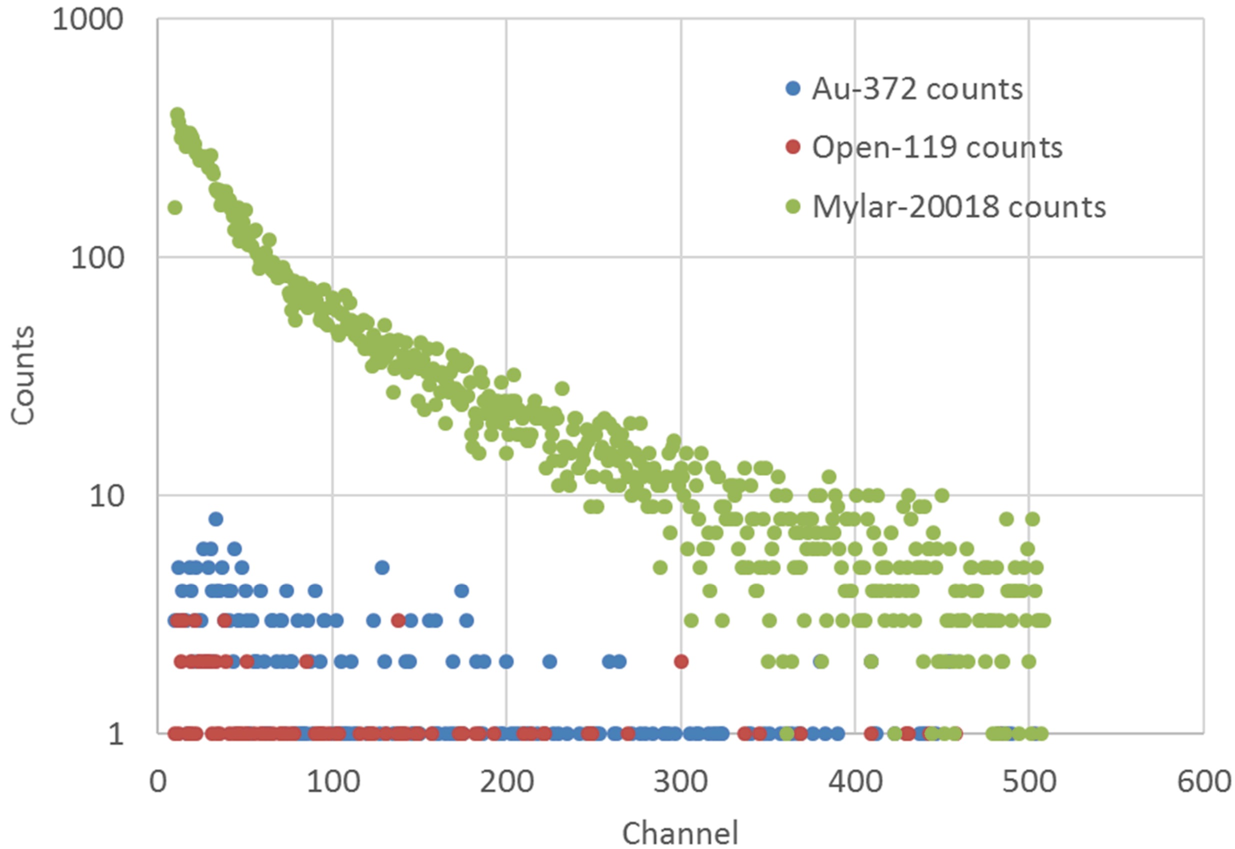

Figure 13: Channeltron counts per channel for a 1 nA beam of 70 keV electrons on 0.5 um Mylar, 0.08 um Au and an open (i.e. no) targets. The labels also include the number of integrated counts.

From the data plotted in Figure 13, the yield of recoiled H atoms for the Mylar target was 66 H/nC. From the theory above we were expecting 0.776 H/nC. This yield was still much larger than expected by a factor of almost 100!

ANTICIPATED OUTCOMES AND IMPACTS:

A detailed literature review revealed the source of this discrepancy. Serkovic Loli et al. [10] found that the stopping power nearly vanishes for ions and atoms on insulators at very low energies. A slightly modified plot from their paper is shown in Fig. 14.

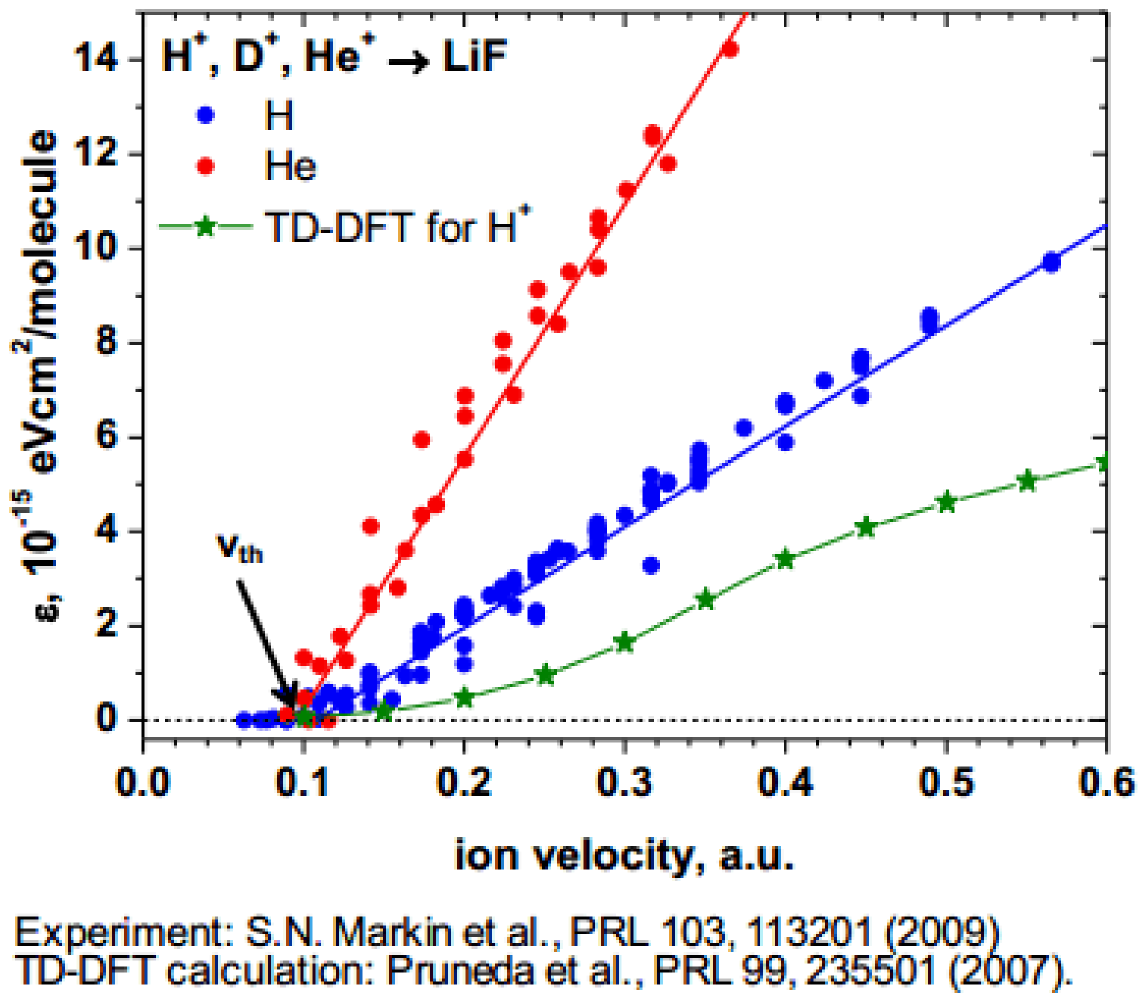

Figure 14: Data and theory of stopping powers of low energy protons in LiF taken from Serkovic Loli et al. [14]. The stopping power nearly vanishes near 0.1 au, which corresponds to a proton energy of 249 eV.

From the Kinematics section above, the maximum energy of H atoms recoiled by 70 keV electrons is 164 eV, which is considerably less than the 249 eV energy where the stopping power of low energy protons nearly vanishes for insulators, like Mylar. In the section on Mylar above, the recoil depth was calculated using the ZBL stopping power theory to be 0.012 microns and a yield to be 0.776 counts/nC, but because of the Serkovic Loli results, this thickness was actually the total thickness of the Mylar, or 0.5 um. Using this thickness of Mylar the yield of detected recoiled H atoms was calculated to be 32.3 H/nC. This theoretical detected yield is now ~1/2 the experimental yield of 66 H/nC. While there could be several reasons for this small discrepancy, the most likely is that the Channeltron efficiency for detecting the low energy H atoms is twice the 10% value used based on the Channelplate results of Crandall [8]. The openings in a Channelplate is 55-65% [11] and this factor of 2 increase in efficiency for a Channeltron as compared to a Channelplate is understandable.

In addition to the 70 keV electrons, the detected recoiled H atom yield was measured at three additional electron energies of 56, 49 and 19 keV. These results are plotted together with theoretical predictions using equation (14) and a detection efficiency of 20% in Fig. 15. The agreement is very good for the highest two energies, but the data is lower than the theory for the lowest two energies. This is probably because a constant 20% detection efficiency was used, and from Fig. 5, it is clear that this efficiency decreases for Channelplates, and probably for Channeltrons, for decreasing H energies.

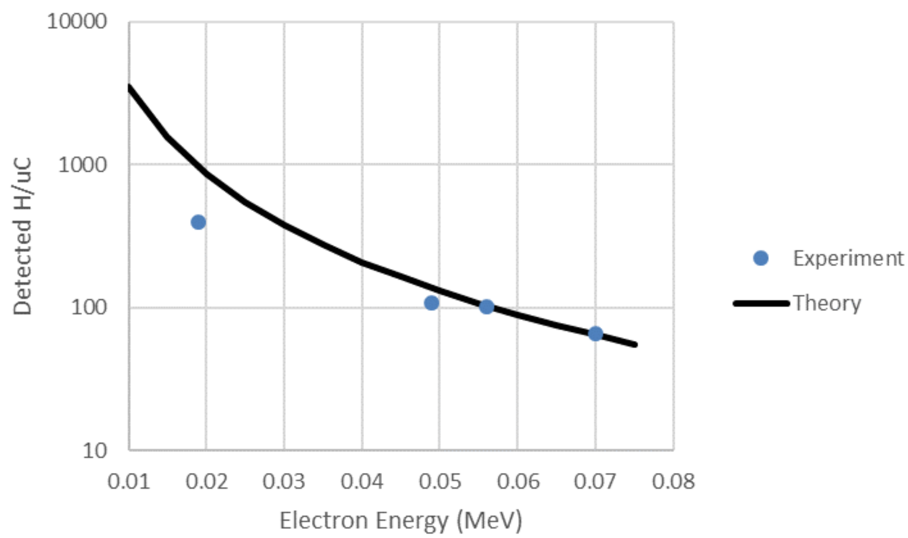

Figure 15: Detected recoiled H yield as function of electron energy. The Channeltron efficiency was assumed constant at 20%, but probably decreased at the lower energies.

The feasibility of using moderate energy electrons to recoil and detect H in electron transparent has therefore been proven by this LDRD.

CONCLUSION:

This LDRD project succeeded in proving the feasibility of using electrons at energies from 19-70 keV to recoil and detect H atoms from thin transmission mounted thin films. This important discovery should lead to the development of electron Elastic Recoil Detection (e-ERD) of H and perhaps He isotopes using the electrons from a Scanning Electron Microscope (SEM) or Transmission Electron Microscope (TEM). With an SEM, the lateral resolution could approach a few nm, while in a TEM perhaps a few Angstroms! This resolution is 1000s of times better than currently available on nuclear microscopes. Such improvements in microscopy are rare, and it is easy to envision how this discovery will benefit H-based materials science and engineering.

For example, there are many measurements that would benefit DoE/NNSA that require this degree of high resolution. These include the distribution of H isotopes (and ^{3}He) in individual grains of materials relevant to TPBARs, neutron generator targets, H and He-embrittlement of weapon and other metallic components, Tritium Sustainment Programs, issues with Gas Transfer Systems, fuel cells, batteries… To address these and many other issues, the potential nm-scale lateral resolution enabled by the successful demonstration of e-ERD reported here and implemented on a future SEM or TEM is required. The development of such a world-unique system would have utility to virtually every hydrogen isotope science topic of interest to the NW complex and H-issues of importance to energy R&D.

The development of an SEM/TEM e-ERD system for nm-resolution mapping of H in materials will therefore be vigorously pursued. A small project to demonstrate e-ERD on a recently purchased SEM in the IBL was funded at the very end of this fiscal year (FY17), and executives in the Enhanced Surveillance Campaign have been contacted requesting support for next fiscal year (FY18).

A presentation of the successful development of e-ERD will be given at the 23rd International Conference on Ion Beam Analysis in Shanghai in October. A paper will result from this publication that will be published in Nuclear Instruments and Methods Part B in 2018. It is therefore very important that Sandia realize the H and He nanoprobe enabled by installing the e-ERD capability into an SEM or better a state of the art TEM early next year.

Built on

LDRD

Laboratory Directed Research and Development

REFERENCES:

1. E. Rutherford F.R.S. (1911) LXXIX. "The scattering of α and β particles by matter and the structure of the atom", Philosophical Magazine, 21:125, 669-688
2. J. W. Corbett, Electron Radiation Damage in Semiconductors and Metals, Academic Press (1966).
3. F. Seitz and D. Turnbull, "Advances in Research and Applications", Solid State Physics, Academic Press (1958-1966)
4. C. G. Darwin, Phil. Mag. [6] vol. 25, 201 (1913)
5. Burle, CHANNELTRON ELECTRON MULTIPLIER HANDBOOK FOR MASS SPECTROMETRY APPLICATIONS, available upon request.
6. SIMION 8.1/8.1, Scientific Instrument Services, Inc. and Idaho National Laboratory, http://simion.com/
7. B.L. Peko and T.M. Stephen, "Absolute detection efficiencies of low energy H, H^-, H^+, H_2^+ and H_3^+ incident on a multichannel plate detector", Nucl. Instr. and Meth in Phys. Research B 171 (2000) 597.
8. D. H. Crandall, J. A. Ray and C. Cisneros, "Channeltron efficiency for counting of H+ and H- at low energy", Review of Scientific Instruments 46 (1975) 562.
9. H.H. Anderson and J. Ziegler, "Hydrogen: Stopping Powers and Ranges in All Elements", Pergamon Press, QC794.6.S8A5 (1977).
10. L. N. Serkovic Loli,E. A. Sanchez, O. Grizzi, and N. R. Arista, "Stopping power of fluorides and semiconductor organic films for low-velocity protons",Phys. Review A81, 022902 (2010).
11. T.Gys, "Micro-channel plates and vacuum detectors", Nucl. Instr. and Meth in Phys. Research, Volume 787 (2015) 254-260.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.