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Using High Energy Electrons for Elastic Recoil Detection of Hydrogen*

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Abstract

Materials that incorporate hydrogen are of great interest for both scientific and technological reasons. The Ion Beam Laboratory at Sandia National Laboratories has developed techniques using micron to mm-size MeV ion beams to recoil H and its isotopes (Elastic Recoil Detection or ERD) that can very accurately make such measurements. However, there are many measurements that would benefit the field of materials science and technology that require much better resolution. To address these and many other issues, we have demonstrated that H can be recoiled through a thin film of Mylar 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 has proven 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 isotopes with nm resolution.

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Keywords electron recoil detection Hydrogen

1. INTRODUCTION

Materials that incorporate hydrogen are of great interest for both scientific and technological reasons. Currently, measurements of H in materials using Ion Beam Analysis (IBA) can have near-nm depth resolution but are limited to micron lateral resolution. This means that there are many measurements that would benefit the field of H-based materials science that cannot be performed. Higher resolution would therefore greatly benefit areas such as H-embrittlement in metals, issues with H fuel cells, H implantation and transmutation in fission and fusion nuclear reactor materials, … To address these and many other issues, the Sandia Ion Beam Laboratory (IBL) has pursued proving the feasibility of using high energy electrons for elastic recoil detection – eERD – of H, that if successful could potentially lead to the utilization of transmission electron microscopes (TEMs) for mapping H and H-isotopes at the nm-scale.

In Section 2. the theory of the electron-nucleus collision is reviewed, and equations useful for predicting the performance of an e-ERD system are given. These equations were central in designing the prototype system that was tested in an existing endstation in the target room of the Tandem accelerator in the IBL. The e-ERD system is described in Section 3. In the Section 4. the first tests of the e-ERD concept involving successful measurements of H in Mylar is given. A summary, recommendations for improvements and a path forward for realizing an e-ERD system on a Sandia TEM are given in the Section 5.

2. REVIEWING THE 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 Solid State Physics 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.

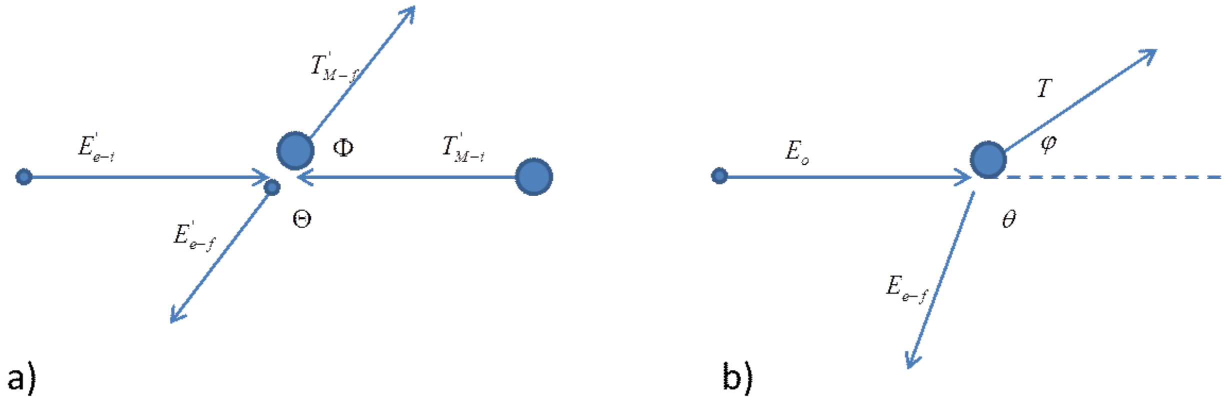

Figure 1: Scattering diagram of electron (small circles) on atomic nucleus (large circles) in a) the center of mass (com) frame and b) the laboratory (lab) frame. The incident and final energies of the electrons are E' and E in the com and lab frame respectively, and T' and T for the nucleus that is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

2.1. Kinematics

For the detailed mathematical derivation of the kinematics of the electron-nucleus interaction the reader is referred to the Corbett book [2]. 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:

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

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:

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

Where as usual

$$\gamma = \frac{1}{\sqrt{1-\beta^2}},\ \text{and}\ \beta = \frac{v}{c} \tag{3}$$

It is also useful to express the recoil energy as:

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

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

2.2 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:

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

The Rutherford cross section then becomes:

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

Darwin later modified Rutherford's cross section for the case of relativistic incident particles deriving the Darwin-Rutherford equation [4]:

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

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:

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

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

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

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

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

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,

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

It becomes important to use the McKinley-Feshback equation 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 for T_{min}/T_{max}=0.99, which as will be shown later corresponds to a lab recoil cone angle of 5.7^o.

2.3 Targets

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, $C_{10}H_8O_4$, $3.51x10^{22}/cm^3$. Mylar also turned out to be ideal because the stopping power of H at energies less than ~250 eV nearly vanishes [5]. This means that all of the H in the film can potentially be recoiled into the channeltron detector. Another target that was 0.08 microns of Au that contained only surface H, and therefore could be used to determine background count rates.

2.4 Calculation of Yield

We can now estimate the signal strength of the recoiled H. The equation for the Yield per second of H recoiled into the e-ERD detector is:

$$Y \approx \phi C_H t\sigma\varepsilon \quad (14)$$

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

$$\phi = 1nA = 6.25x10^{9}\, e/s$$
$$C_H = 3.5x10^{22}\, H/cm^3$$
$$t = 5.0x10^{-5}\, cm$$
$$\sigma = 2.9x10^{-26}\, cm^2$$
$$\varepsilon = 0.1$$

This gives a detected H rate of 32 H/s or 32 H/nC.

3. 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 an existing 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.

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 a small "Bud" box as shown in Figure 2. 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 passed through a region of 4 kG magnetic field provided by permanent magnets and into 3mm diameter "Aperture 1". They then entered "Chamber 2" into a 18mm region that had a 2kG magnetic field also produced by permanent magnets. The recoiled H atoms then passed through "Aperture 2" of 6mm in diameter, but the electrons by this tune were prevented from passing through this final aperture due to the strong magnetic fields in Chambers 1 and 2. The final detector that was selected was a commercial Channeltron electron multiplier [6]. The trajectories of the electrons (red), neutral H atoms (black) and positive H^+ ions (blue) were modeled using the SIMION program [7]. These calculations show that the magnetic fields were 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 [8,9], which are quite similar. Data from Reference 7 indicates that for H atoms in the 100 eV energy range the efficiency is ~ 10%.

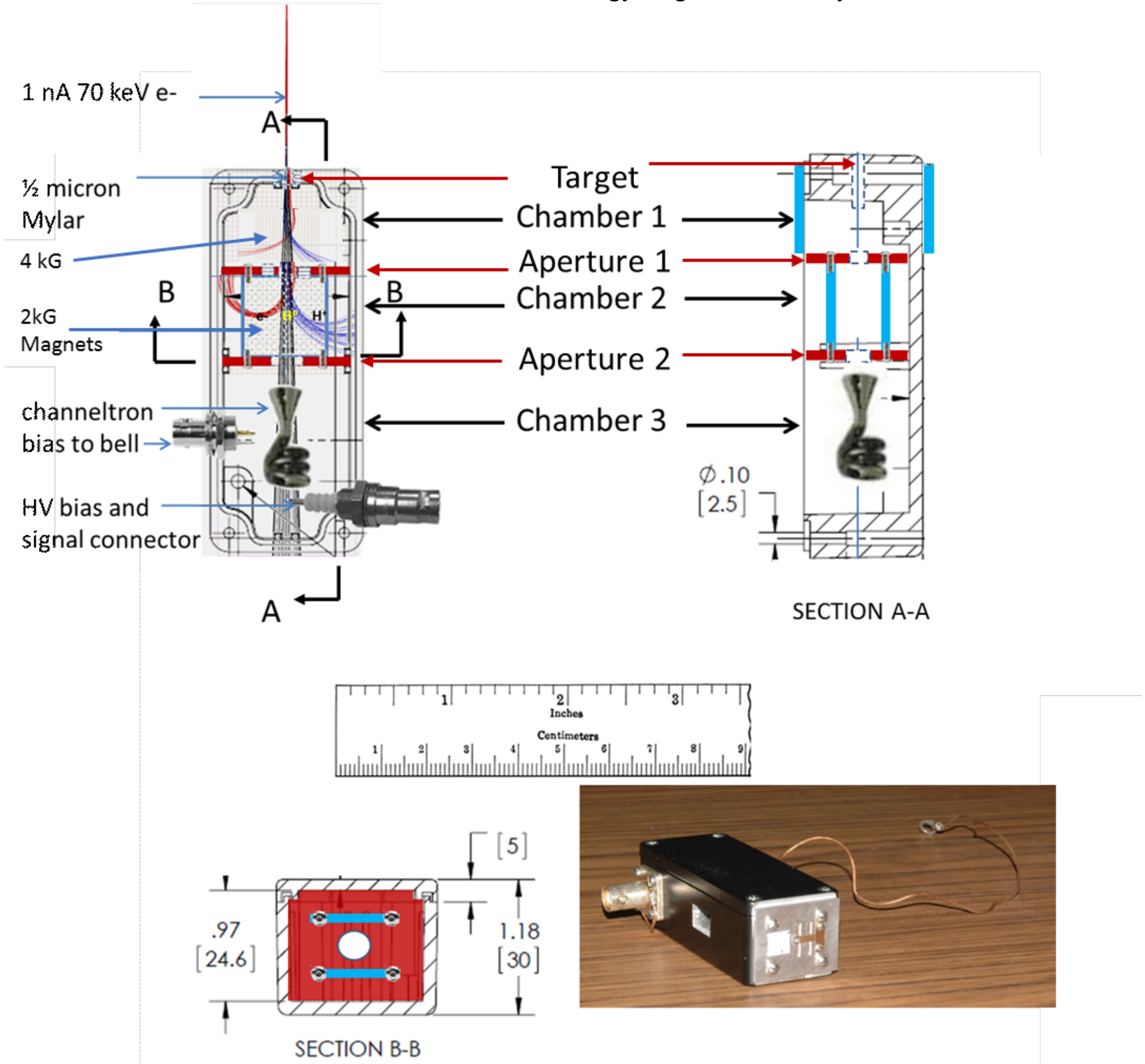

Figure 2: Design drawings of the e-ERD target holder and detector. The various components of the detector are indicated in the upper right, and the trajectories of the electrons (red), neutral H atoms (black) and positive H^+ ions (blue) passing through Chamber 2 were modeled using the SIMION program.

SIMION calculations including simulated scattering from both the target and aperture 1 are shown in Figure 3, and showed that electrons could find trajectories that could pass through aperture 2 into the Channeltron. The solution to this problem was found by adding the 4kG magnetic field in Chamber 1. 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.

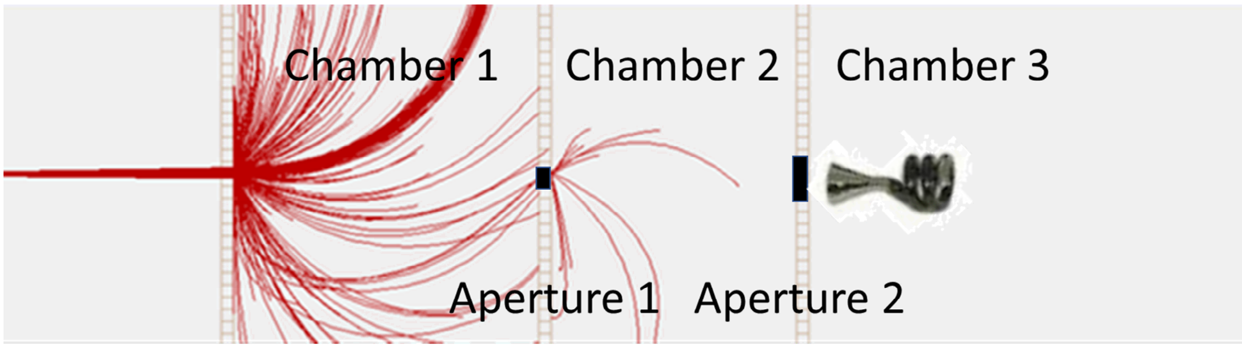

Figure 3: SIMION calculations electron trajectories for three situations of the e-ERD detector system with 1.9 kG magnetic field in Chamber 2 and a 4 kG field in Chamber 1 including electron scattering.

Standard nuclear-based electronics was used to process the signals made when H^o atoms struck the Channeltron. The bell was biased at -300V and the signal end biased at +1800V 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

3. RESULTS

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 Channeltron signal for the e-ERD detector system with magnetic fields in both Chambers 1 and 2 is plotted in Fig. 4 for 70 keV electrons on the Mylar, Au and open targets. The backgrounds in this case have been nearly eliminated by the two sets of magnets, 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.

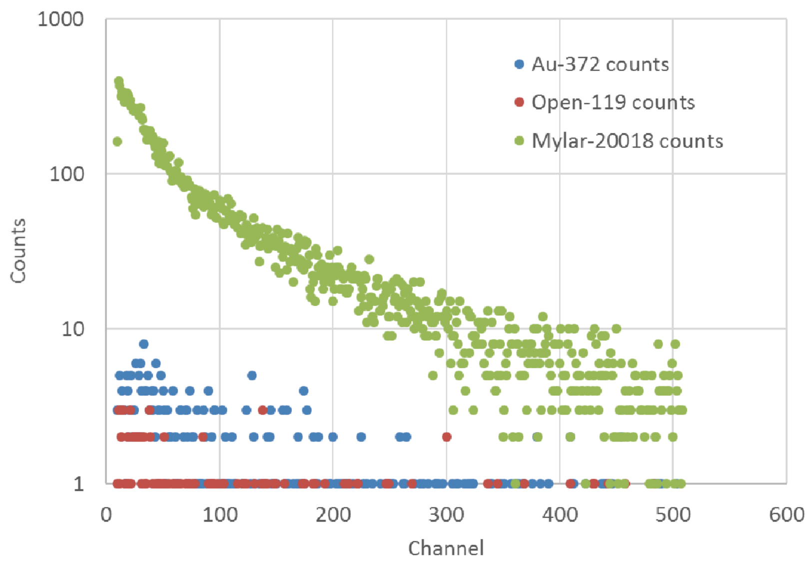

Figure 4: Channeltron counts per channel for a 1 nA beam of 70 keV electrons on 0.5 um Mylar, 0.08 um Au and an open (i.e. no) targets. The labels also include the number of integrated counts.

From the data plotted in Figure 4, the yield of recoiled H atoms for the Mylar target was 66 H/nC.

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 ~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 [9]. The openings in a Channelplate is 55-65% [10] 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. 5. 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 this efficiency decreases in Channeltrons for decreasing H energies.

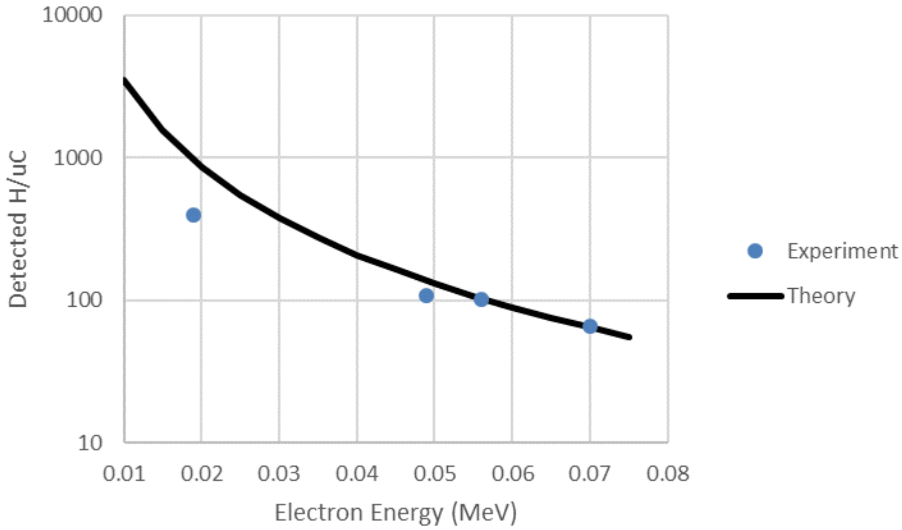

Figure 5: Detected recoiled H yield as function of electron energy. The Channeltron efficiency was assumed constant at 20%, but probably decreased at the lower energies.

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

4. CONCLUSIONS

This 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.

5. REFERENCES

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FIGURES AND CAPTIONS (SAME AS IN TEXT ABOVE)

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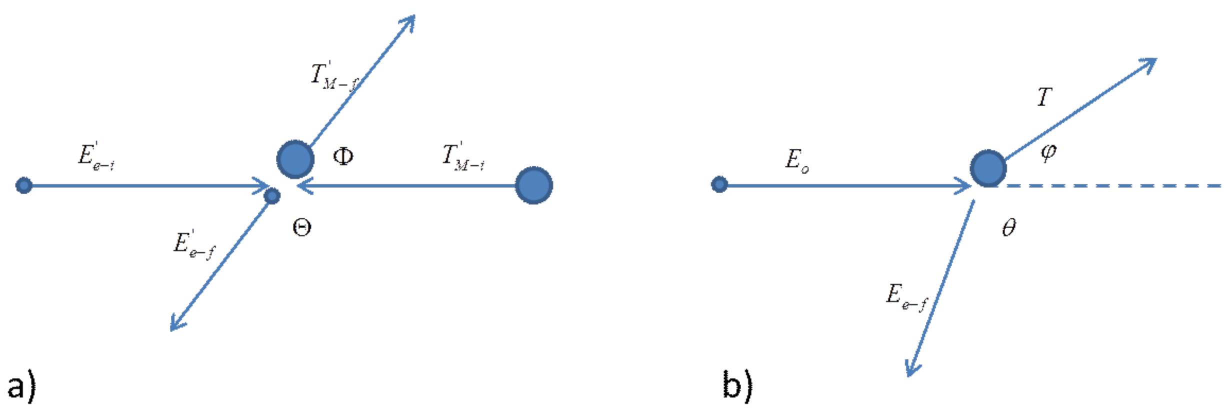

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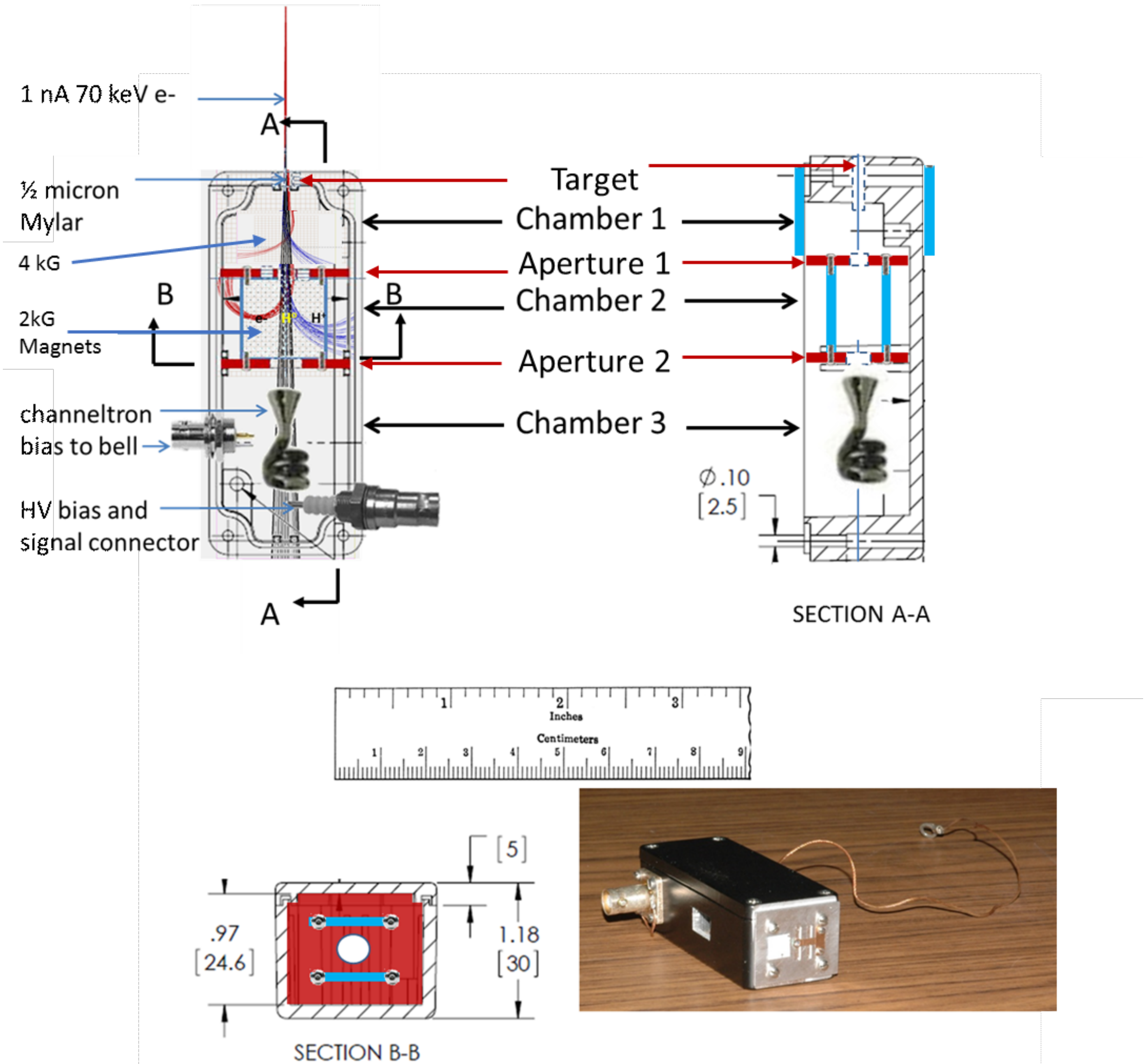

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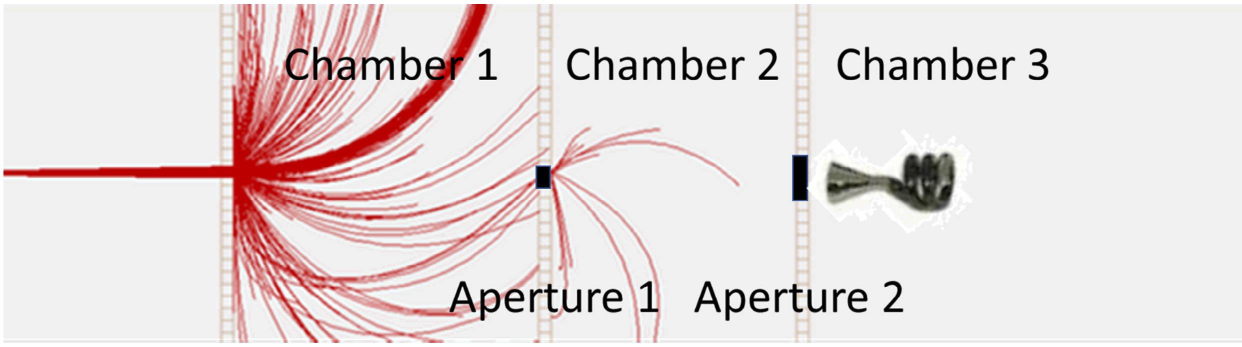

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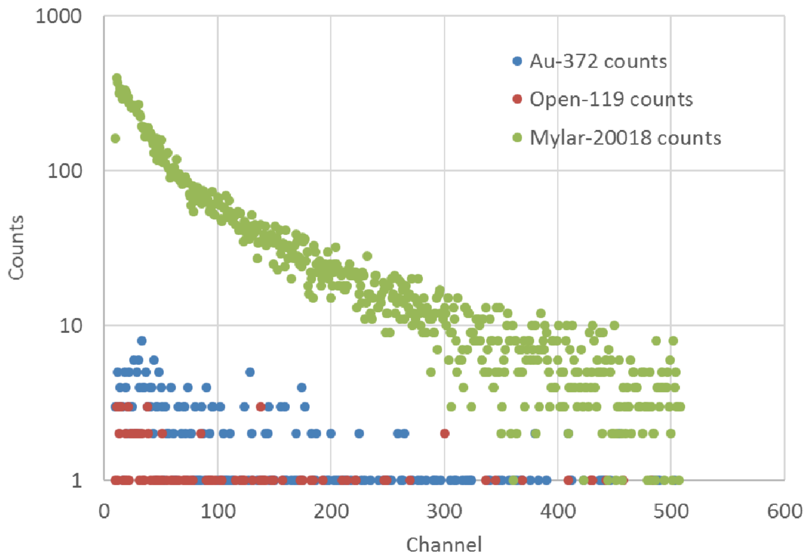

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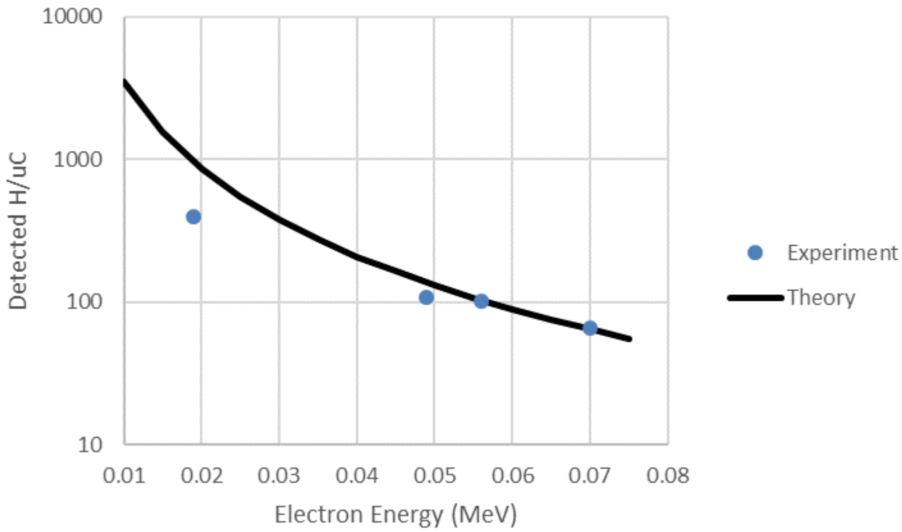

Figure 10: Detected recoiled H yield as function of electron energy. The Channeltron efficiency was assumed constant at 20%, but probably decreased at the lower energies.