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Using High Energy Electrons for Elastic Recoil Detection of Hydrogen – eERD

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Elastic Recoil Detection (ERD) was first used in the IBL at Sandia in the late 70’s using high energy heavy ions like 28 MeV Si^{5+}

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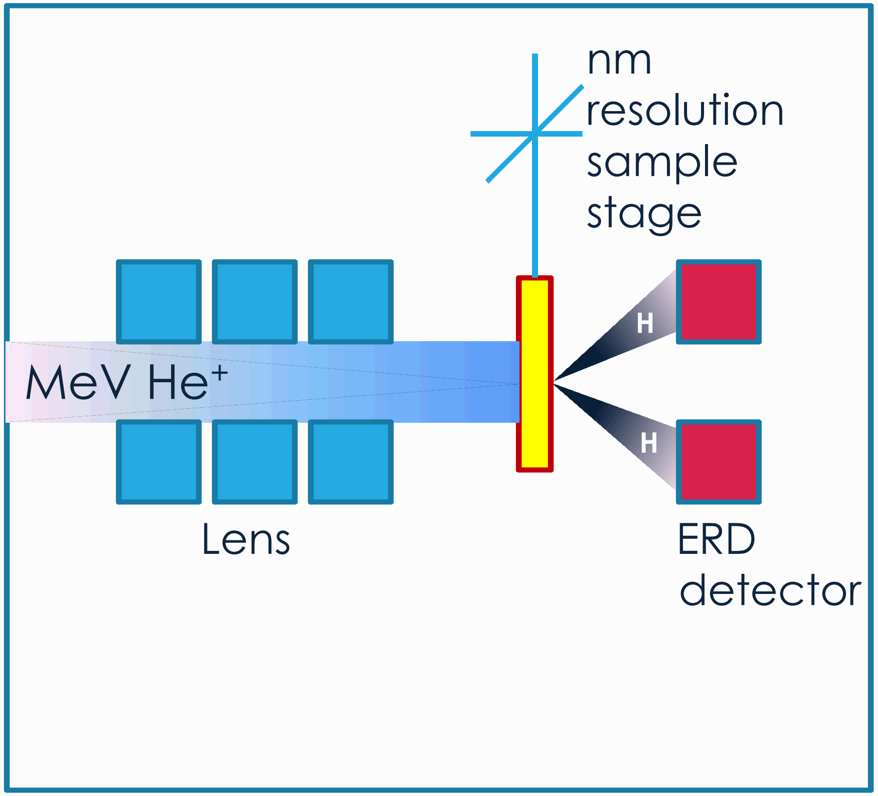
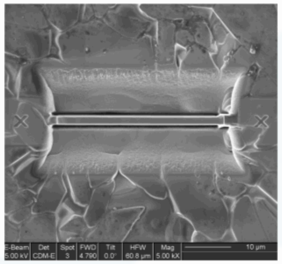
- …but soon after that we discovered that relatively low energy (~1 MeV) He^{+} could also be used for profiling all the H isotopes.
- …and this led to a nuclear micro-probe system for H profiling in 3D on the Pelletron
  - …and this worked
  - but the lateral resolution was >1 micron

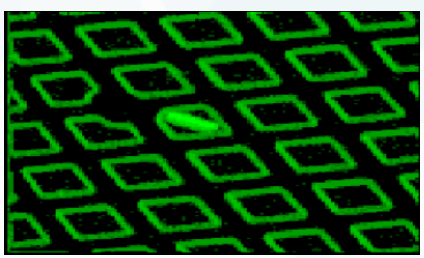


Ceramic Samples analyzed with µERD to determine where H (i.e. H_2O) is located



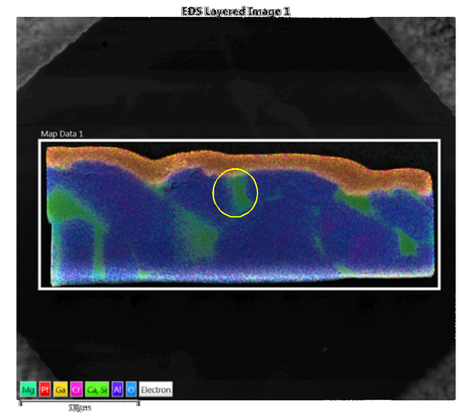
FIB sample prior to lift-out

Sample mounted onto TEM grid and measured with .75 MeV He^+ Rutherford Forward Scattering

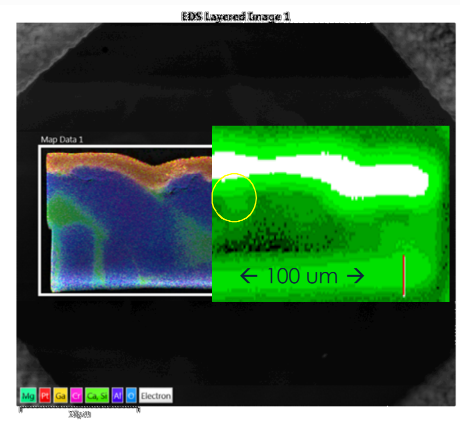


- Sample was 94% alumina + 6% intergranular glassy phase
- Glassy phase on surface appears to impact wetting/adhesion of epoxy potting and wetting of active braze alloy in braze joints
- Depletion of glassy phase on surface correlated with differences in H_2O concentration in process ambient at vendor and SNL
- H_2O well known to modify voltilization behavior of common commercial glasses
- ***H is found in the SiO_2 glassy phase***
- Very little H found in another sample processed with little H_2O
- Agreed with IR
- …but the ERD mapping of H would have obviously benefited if the latteral resolution was as good as that of the SEM.

SEM



µERD



This year we started a project to demonstrate the feasibility of using 70 keV electrons for ERD analysis of H and He isotopes.

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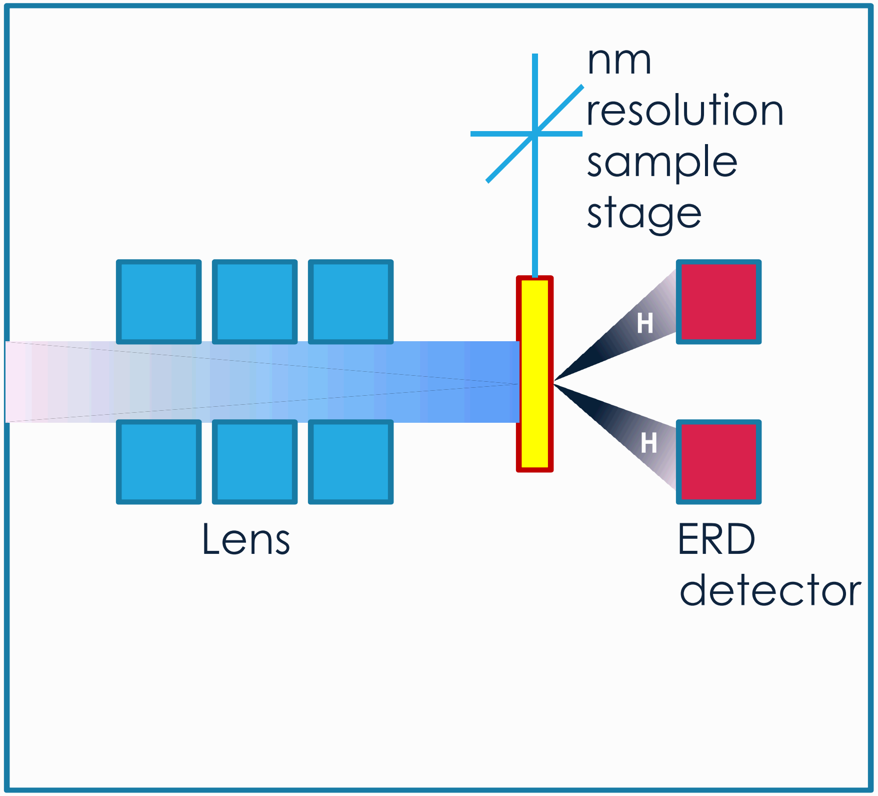
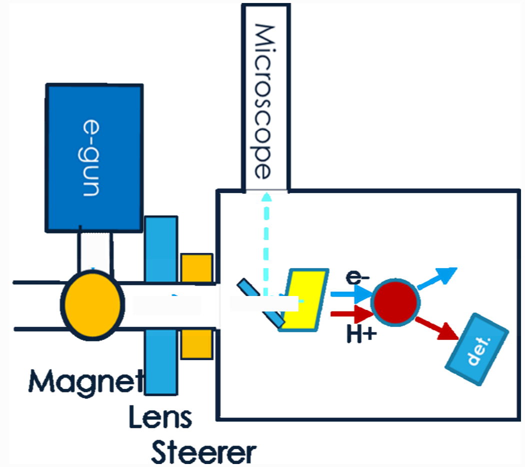
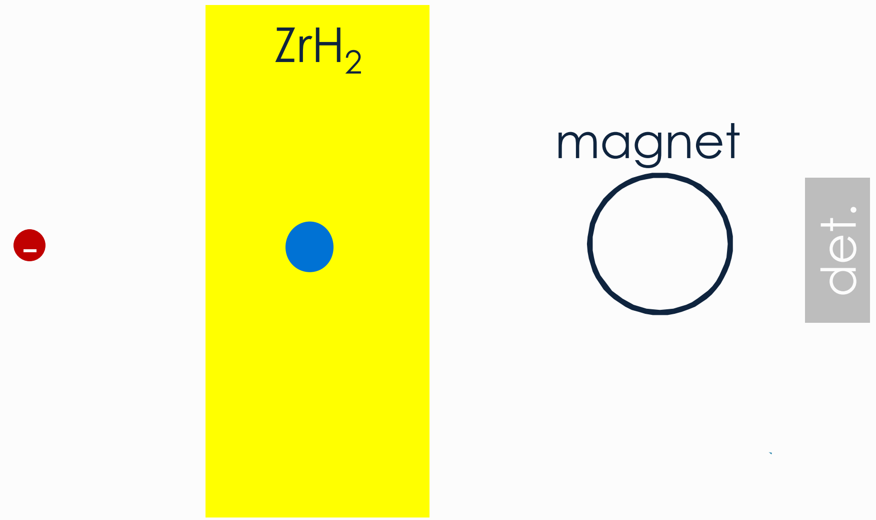
- ...but soon after that we discovered that relatively low energy (~1 MeV) He^+ could also be used for profiling all the H isotopes.
- ...and a nuclear micro-probe system for H profiling in 3D on the Pelletron
  - ...and this worked
  - but the lateral resolution was only ~ 1 micron
- This project explored whether even lighter projectiles, i.e. electrons, can also be used for ERD of H isotopes?
  - ...and on a TEM with nm resolution?



The feasibility experiment for eERD

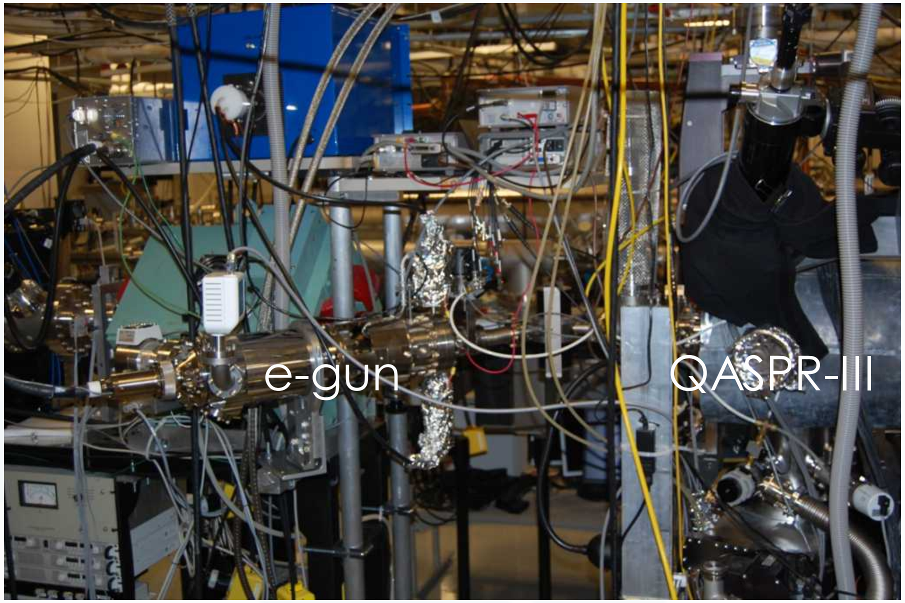
1. 70 keV e's obtained from egun on QASPR-III
2. Deflected by magnet into endstation
3. Focused to ~mm onto H-containing thin film target
4. As e's penetrate target they loose little energy
5. …but can recoil H atoms
6. e's are transmitted, and H atoms are recoiled out of the film
7. Both go through a magnet that
    1. bends the e's away from the detector
    2. transmits the H atoms to a detector
8. The yield of detected H atoms is converted to the concentration of H atoms in the sample

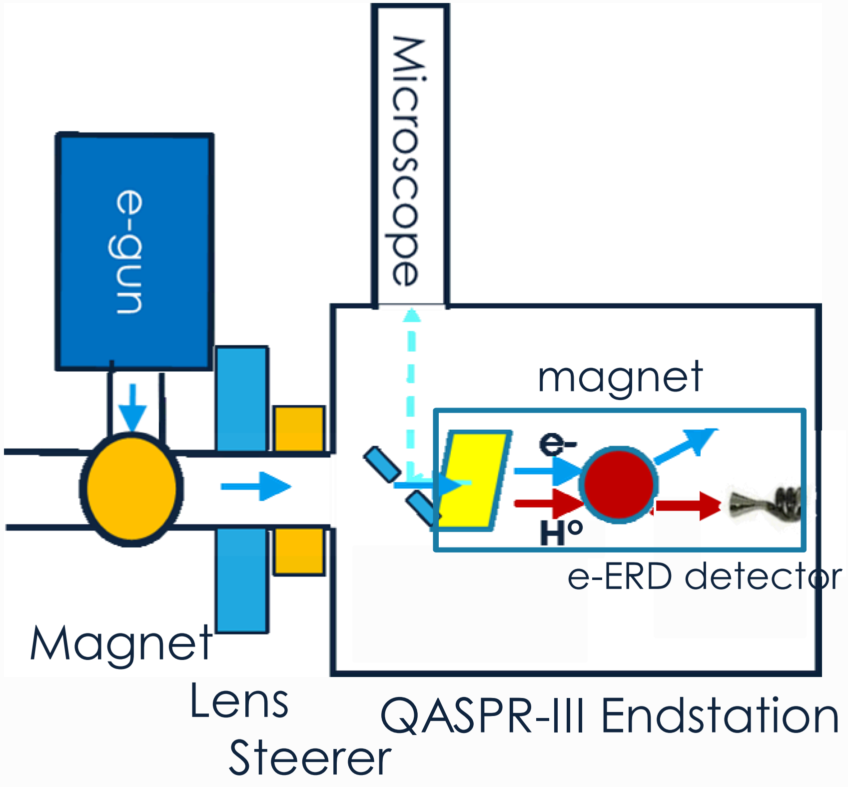




Proposed R&D

To review, we wanted to demonstrate that H (probably neutral H atoms) can be recoiled from a thin H-containing sample by 70 keV electrons on the QASPR-III endstation, separated from the transmitted electron beam, and detected with fairly high efficiency.





The energy for H recoiled at 0° is given by:

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

$$\gamma = \frac{1}{\sqrt{1-\beta^2}} = 1 + \frac{T_e}{mc^2}, \qquad \beta = \frac{v}{c} = \sqrt{1-\frac{1}{\gamma^2}},$$

$$T_e = kinetic\ energy\ of\ electron, \qquad m = mass\ of\ electron, \qquad M = mass\ of\ proton$$

for 70 keV e's

gamma=	1.137
v/c=	0.476

m=	0.000544662	amu
M=	1	amu
Te=	0.07	MeV
Tmax=	164	eV

What's the recoil event probability or cross section?

The relativistic quantum mechanical cross section for an electron to recoil a proton is given by the McKinley-Feshbach equation:

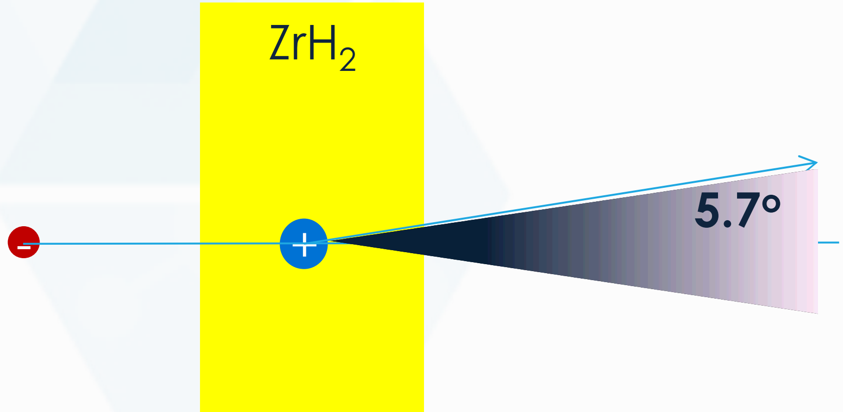
$$\sigma_{McF} = \pi\left(\frac{Ze^2}{mc^2}\right)^2 \frac{1}{\beta^4\gamma^2}\left[\left(\frac{T_{max}}{T_{min}} - 1\right) - \beta^2 \ln\left(\frac{T_{max}}{T_{min}}\right) + \pi\alpha\beta\left\{2\left[\left(\frac{T_{max}}{T_{min}}\right)^{1/2} - 1\right] - \ln\left(\frac{T_{max}}{T_{min}}\right)\right\}\right]$$

$$where\ T_{min} = \frac{2p_o^2 cos^2(\varphi)}{M}\ and\ \varphi\ is\ the\ maximum\ recoil\ angle$$

for the case of 70 keV e's recoiling protons

gamma=	1.137	
v/c=	0.476	
alpha=	0.007	
Tmin/Tmax	0.990	
Tmax=	164.211	eV
Tmin=	162.568	eV
phi=	5.740	deg

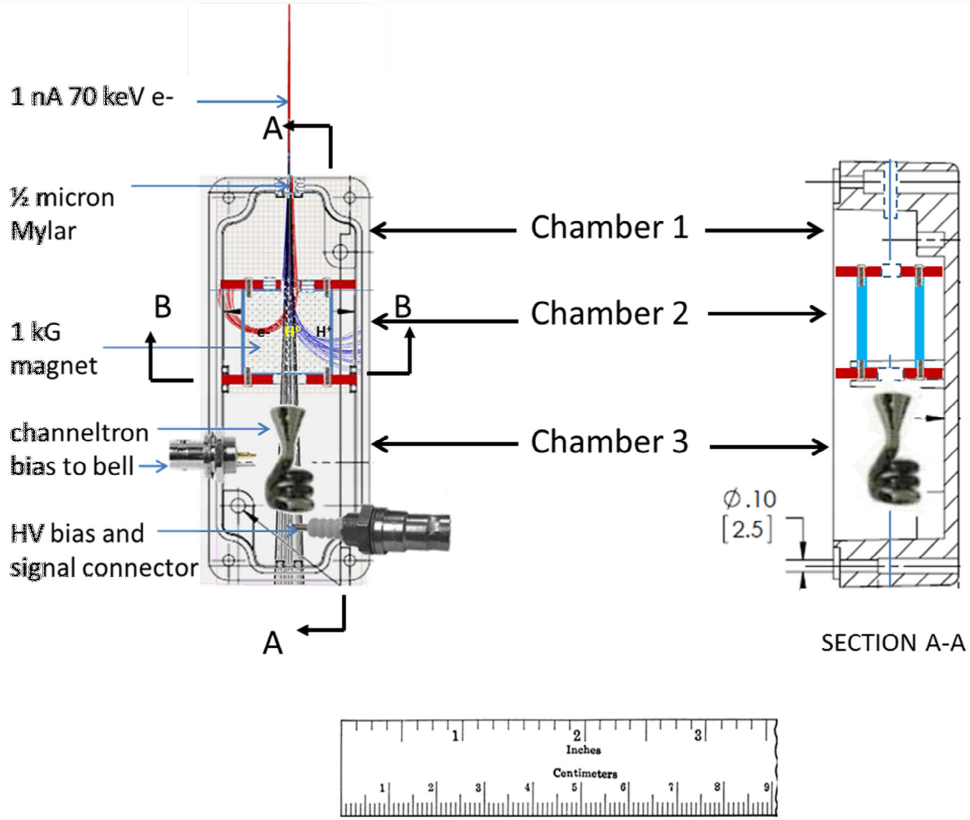
XSo=	376.282	fm2
XS=	3.801	fm2
XS=	2.945	fm2
		MF
XS=	2.94E-26	cm2
	0.029	barns
	29.4	mbarns

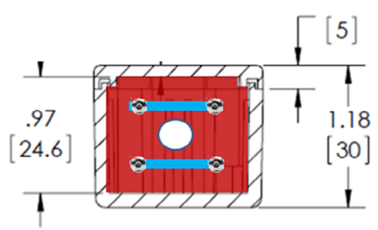


E. Rutherford F.R.S. (1911) LXXIX. "The scattering of α and β particles by matter and the structure of the atom", Philosophical Magazine, 21:125, 669-688

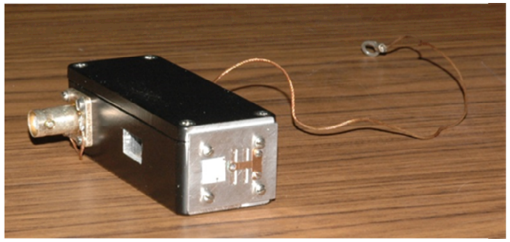
XS Ruth. = 33.2 mbarns

Mark 1 e-ERD system

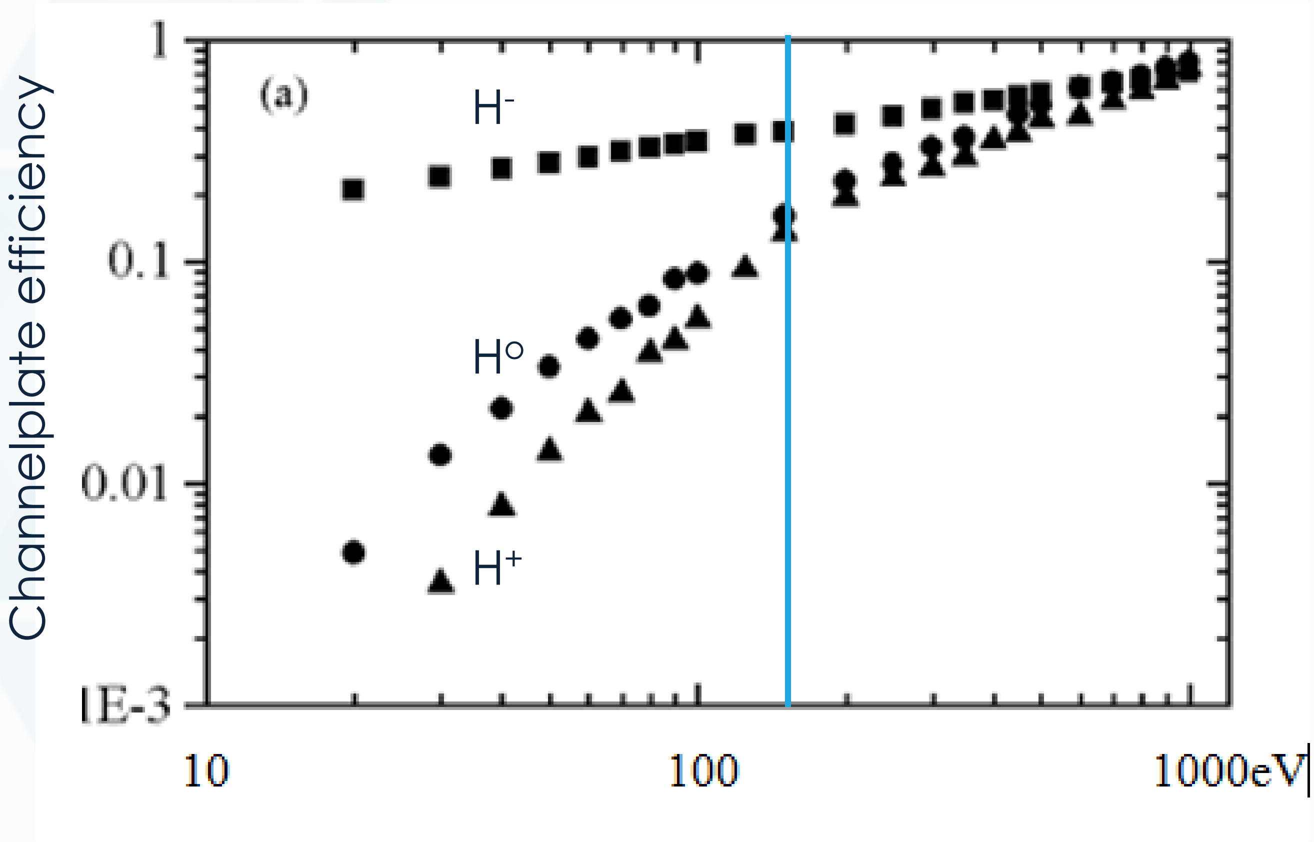




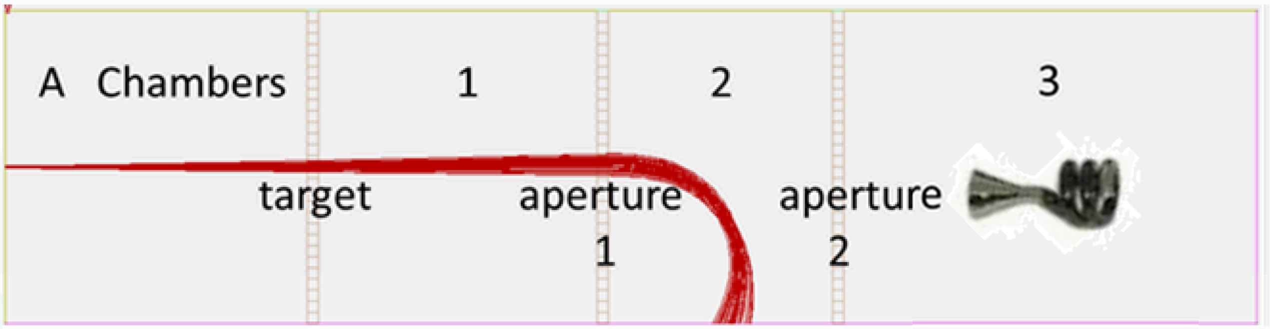
SECTION B-B



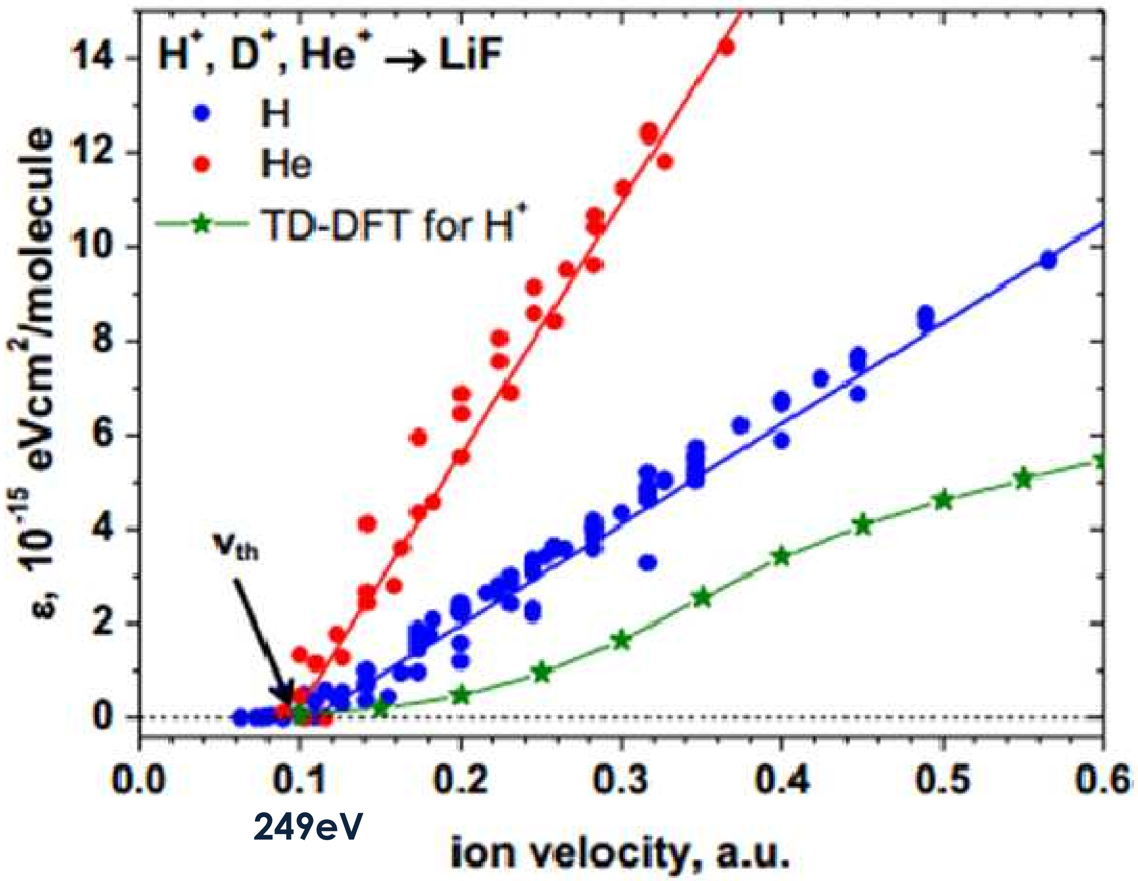
Detection efficiency of a Channelplate for low energy H^- (squares), H^o (circles), and H^+. The energy of the recoiled H^o in this experiment is 164eV, and the efficiency is therefore ~10%.



SIMION calculations indicate 2 1kG magnets adequate to deflect 70 keV electrons away from channeltron in chamber 3



½ micron Mylar $C_{10}H_8O_4$ was selected to be the initial target because of its high H concentration 3.5×10^{22} H/cm^3 and because the stopping power of low energy H in an insulator like Mylar is nearly ZERO!



Experiment: S.N. Markin et al., PRL 103, 113201 (2009)
TD-DFT calculation: Pruneda et al., PRL 99, 235501 (2007).

Based on that cross section, what is the count rate we would expect when the target is Mylar?

The equation for the Yield per second of H recoiled into the e-ERD detector is:

$$Y \approx \phi C_H t \sigma \varepsilon$$

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

$\phi = 1nA = 6.25x10^9 e/s$

$C_H = 3.5x10^{22} H/cm^3$

$t = 5.0x10^{-5} cm$

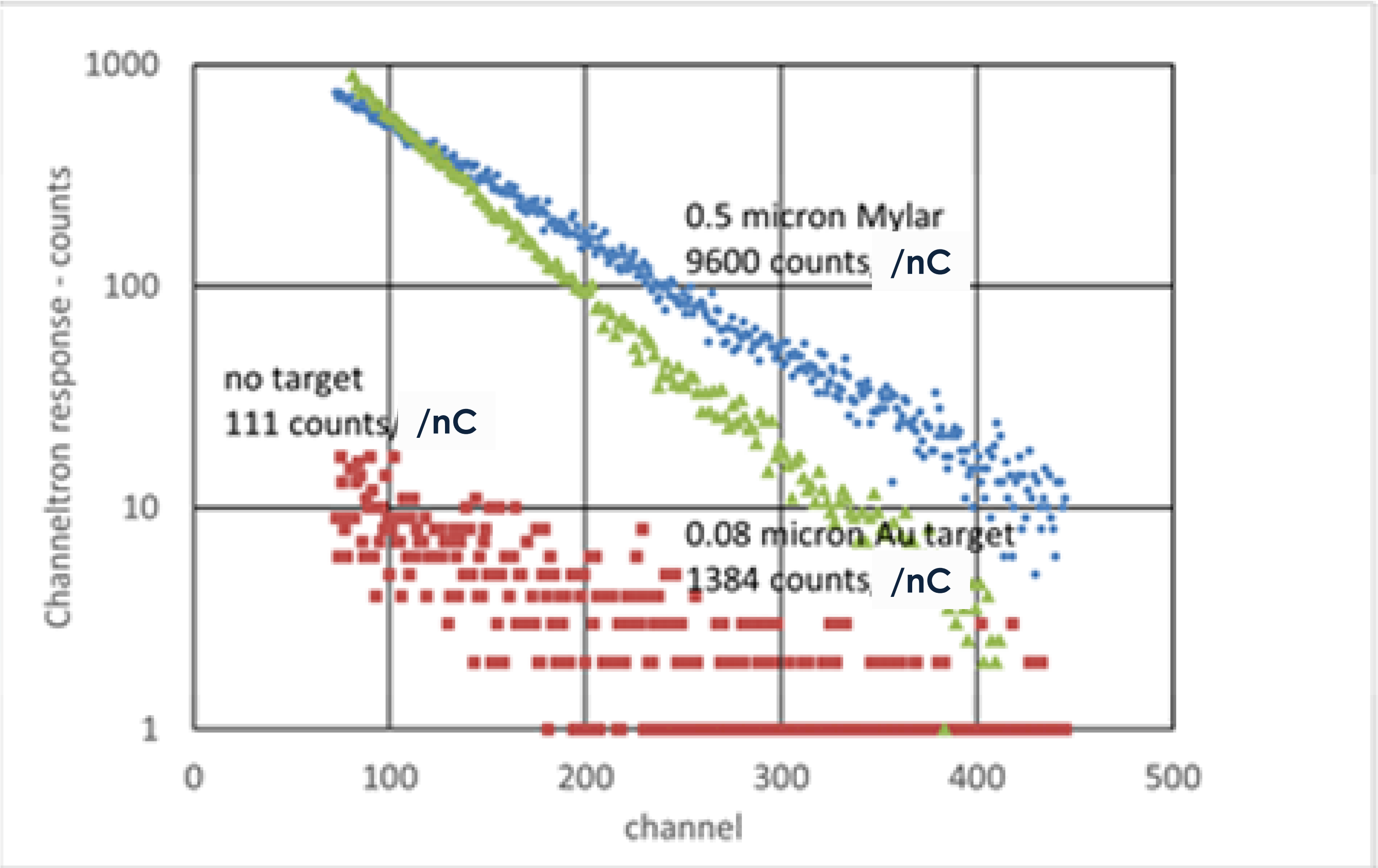
$\sigma = 2.9x10^{-26} cm^2$

$\varepsilon = 0.1$

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

...and these H atoms would have energies of 164 eV.

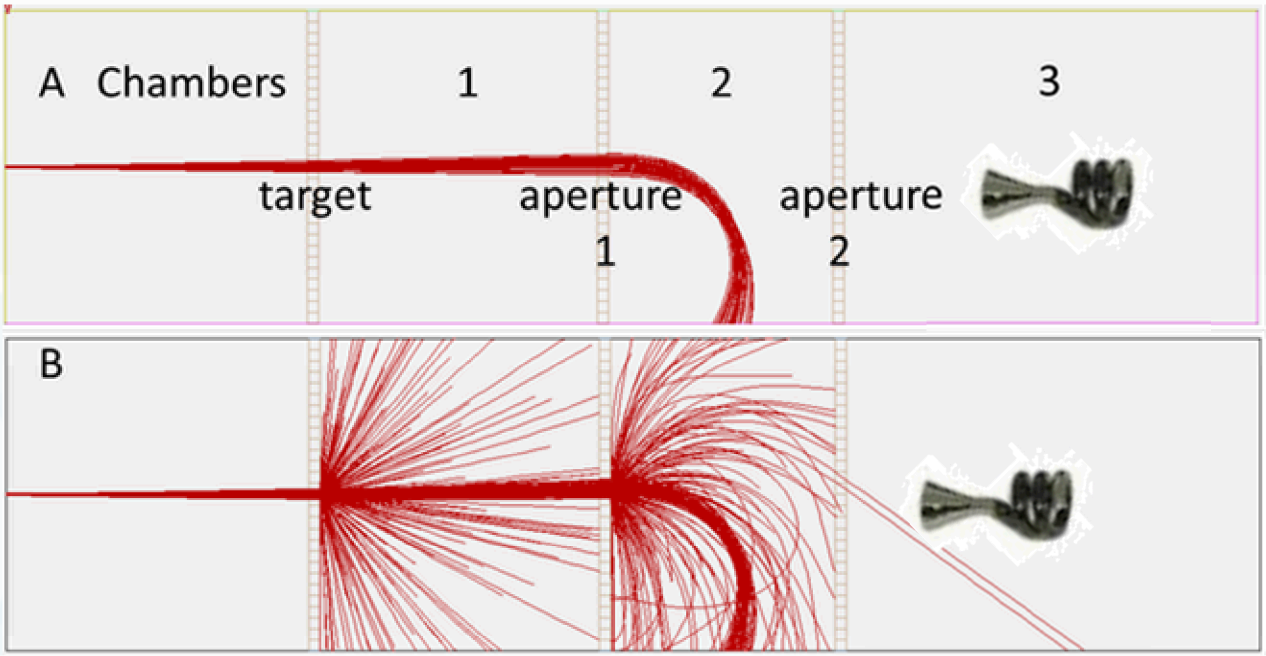
Amplified Channeltron signal output for 1nA of 70 keV electrons placed on 0.5 um Mylar foil (blue), the 0.08 um Au foil (green) and with no target (red) for 50 seconds.



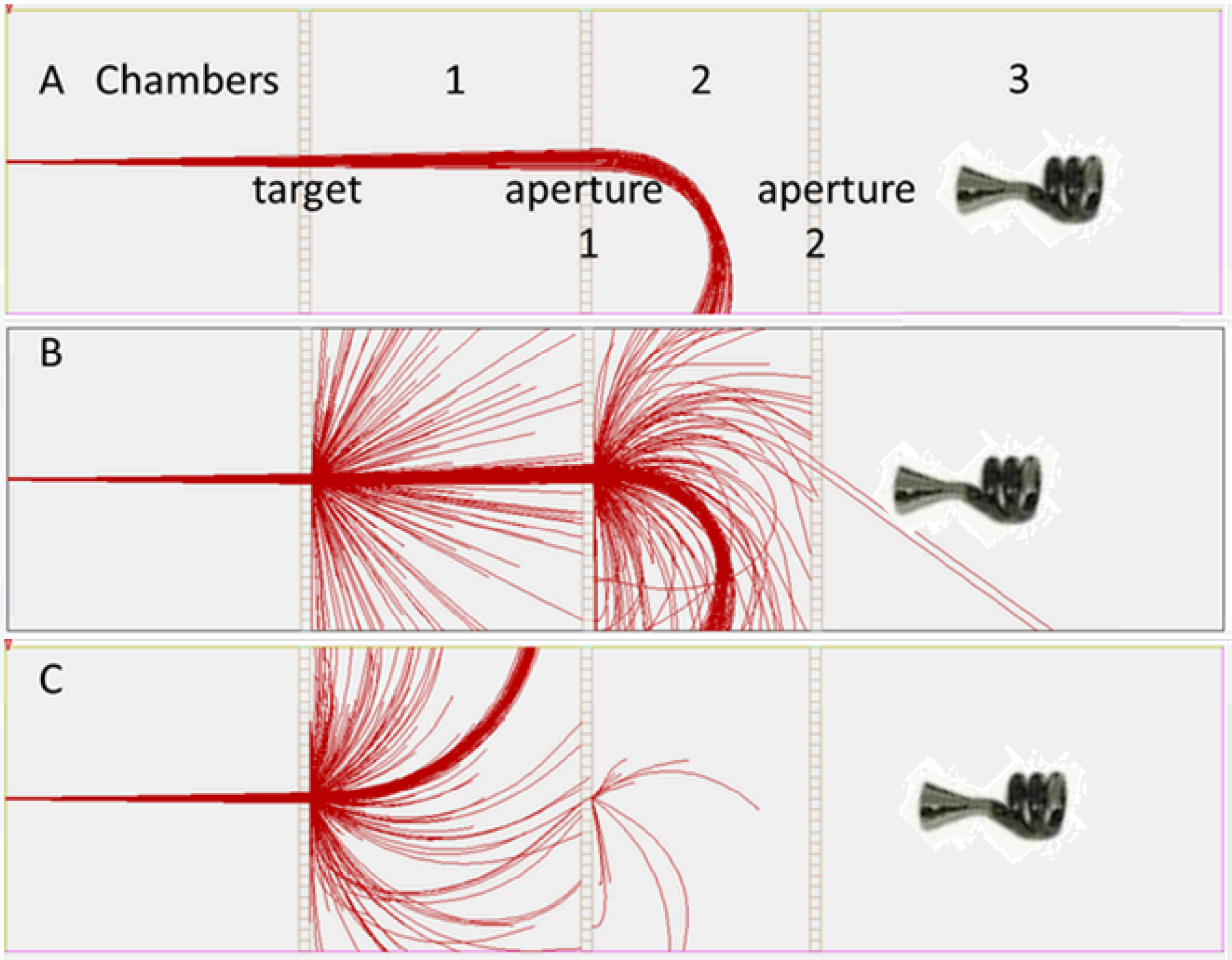
Count rate way to high☹

- From the theory presented above, the yield of H atoms recoiled from the 0.5 micron Mylar foil was expected to be 32 H/nC and nearly 0.0 for the Au foil and open target. The yields plotted the previous figure 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 indicate 2kG magnets are inadequate to deflect 70 keV electrons that scatter from the target and aperture 1 and can pass through aperture 2 to the channeltron in chamber 3



SIMION calculations indicate that adding 4 kG magnets to chamber 1 will nearly eliminate electron scattering from aperture 1 and prevent electrons from hitting the channeltron



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.

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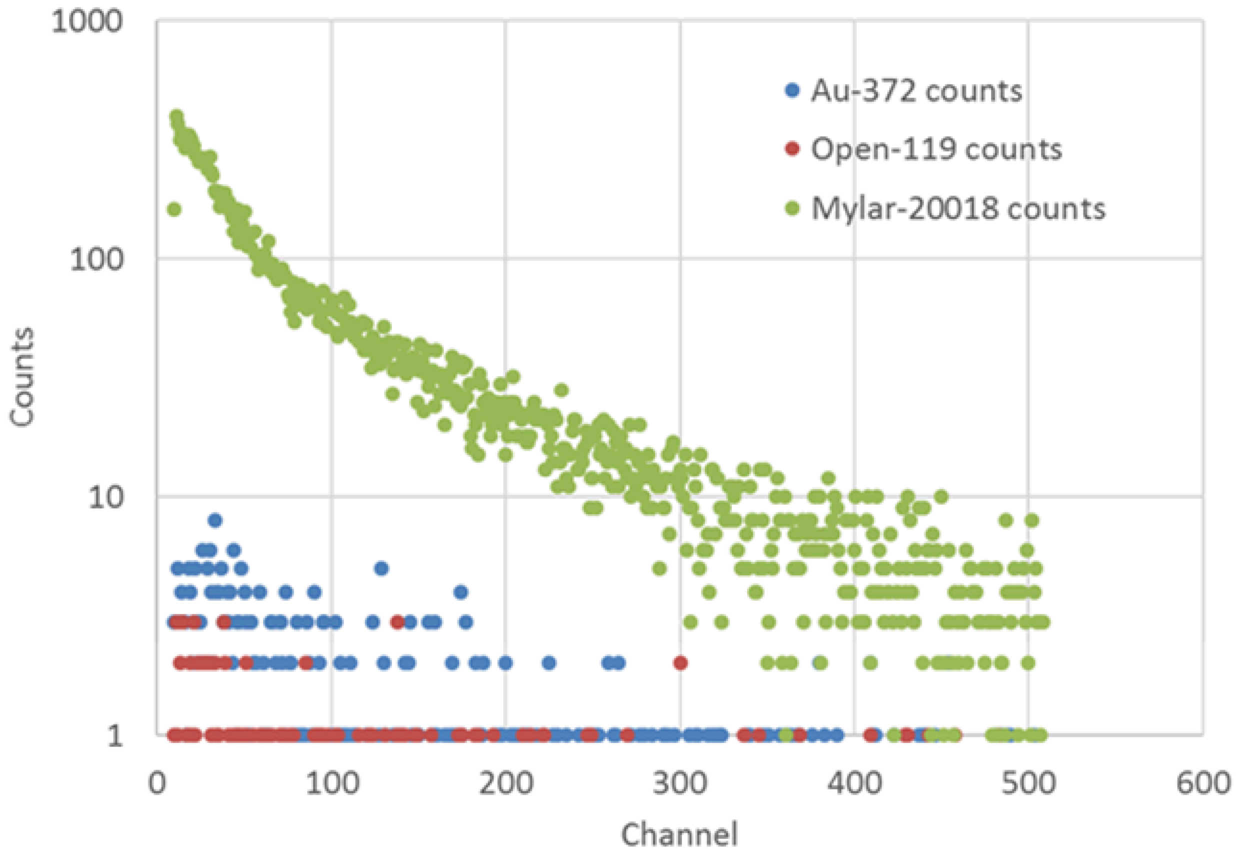
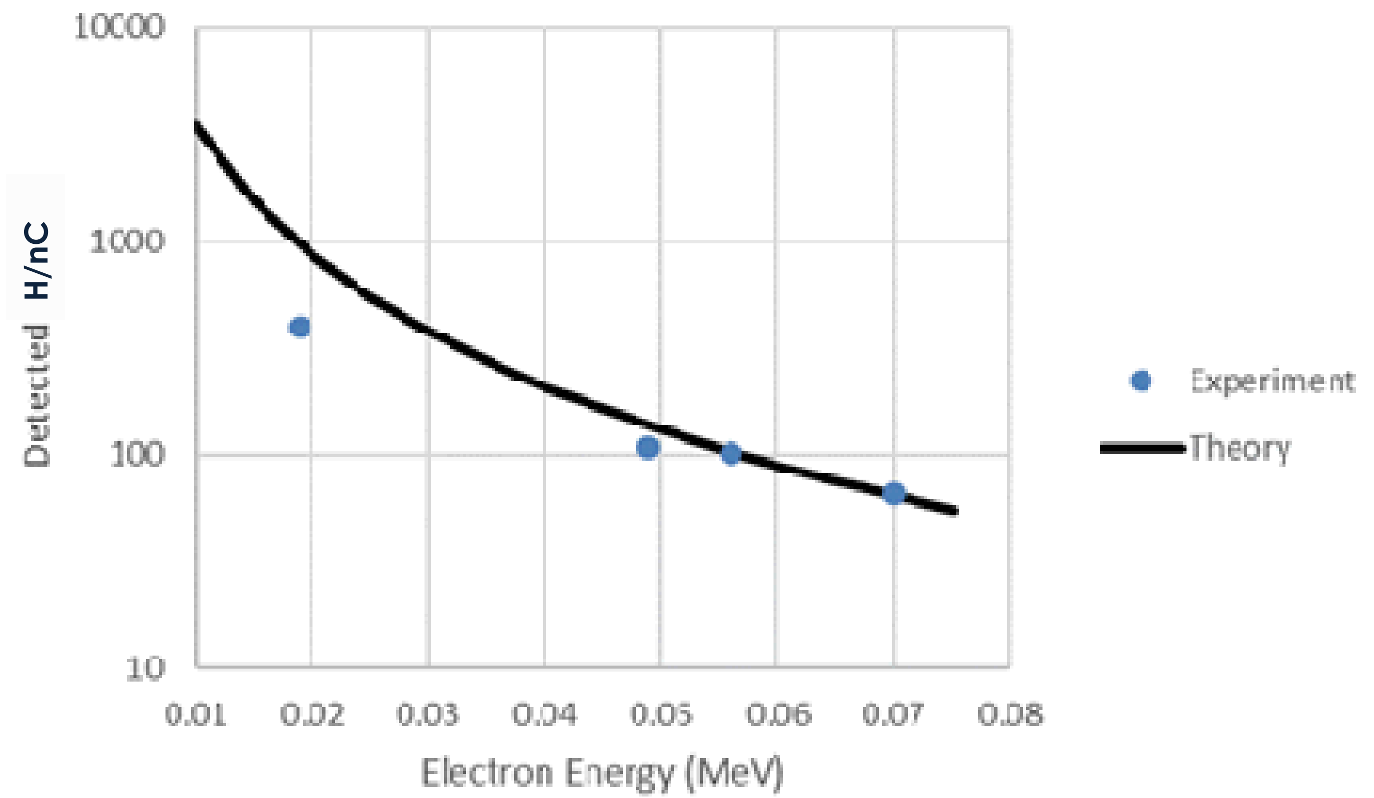


The yield of recoiled H atoms for the Mylar target was 66 H/nC. From the theory above we were expecting 32 H/nC. So this count rate was only high by a factor of 2, which can be explained by a higher efficiency of the Channeltron.

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



The project was successful☺

- 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 insulator 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.