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Matrix Isolation Spectroscopy Applied to Positron Moderation in Cryogenic Solids

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AIR FORCE RESEARCH LABORATORY, MUNITIONS DIRECTORATE

Air Force Materiel Command ■ United States Air Force ■ Eglin Air Force Base

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14. ABSTRACT

We report results of Matrix Isolation Spectroscopy (MIS) experiments performed on working cryogenic rare gas solid (RGS) positron (e^+) moderators. The e^+ is the antiparticle to the electron,[1-3] and positrons are produced by energetic processes that result in very broad kinetic energy distributions (KEDs) - typically spanning hundreds of thousands of electron volts (eV). Trapping and manipulating e^+ with electromagnetic fields requires narrowing these KEDs below a few eV, which is accomplished *via* velocity-dependent interactions in a normal matter "moderator." A fast e^+ entering a wide bandgap dielectric RGS moderator slows rapidly within the first ~ picosecond, producing a track of ionized and electronically excited species. However, once KE_{e+} drops below the ~ 10 eV minimum required for generating such excitations, these interactions abruptly switch off, and the e^+ enters a phase of "hyperthermal diffusion" that can last for nanoseconds. Positrons that reach a free surface of the moderator before annihilating with an electron may escape into vacuum where they can be manipulated. The best known e^+ moderator is cryogenic solid Ne, which still only delivers efficiencies < 1 %; the other 99+ % of the nascent fast e^+ are wasted. Additionally, the RGS moderator efficiency is known to decrease during operation, which is attributed variously to the buildup of radiation damage, and/or to contamination of the moderator surface by residual gas deposition.

We constructed a novel apparatus that permits optical access to a working cryogenic solid moderator. Our original motivation was to test our hypothesis that solid parahydrogen (pH_2) should be an even better e^+ moderator than solid Ne, while simultaneously monitoring the condition of the moderators by infrared (IR) absorption spectroscopy. Unfortunately, the performance of our ortho/para hydrogen converter (o/p converter) has deteriorated and we only achieved residual orthohydrogen (oH_2) concentrations ~ 1000 ppm in these experiments. While we successfully observed the production of slow e^+ from Ne, Ar, and Kr moderators (background corrected count rates ~ 10 Hz), our oH_2/pH_2 experiments failed to yield a statistically significant slow e^+ signal. Indeed, a ≈ 1 μm thick oH_2/pH_2 overcoat was sufficient to kill the slow e^+ signal from a working solid Ne moderator. We performed several experiments with deliberately doped (CO and H_2O) RGS moderators, and with RGS moderators overcoated with solid CO and water ice, in which we measured quantitatively the dopant concentrations and the thicknesses of the moderator and overcoat layers from the IR spectroscopic data. Our results indicate that the impact of H_2O impurities on RGS moderator efficiency has been overestimated in the literature, and point instead to residual H_2 gas as a possible culprit.

15. SUBJECT TERMS
Positron moderation, rare gas solid, matrix isolation, parahydrogen

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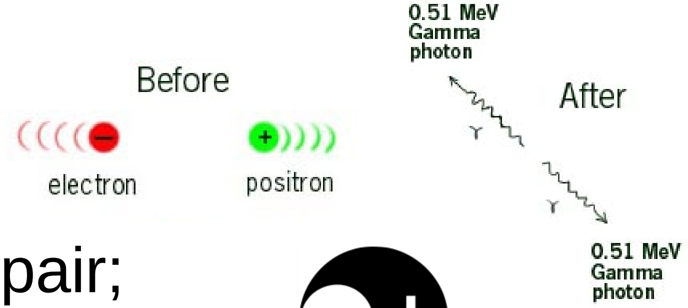
MATRIX2011
Physics & Chemistry of Matrix Isolated Species
U. British Columbia, Vancouver, Canada, 10-15 Jul 11

Outline

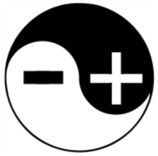
- Positrons introduction
- Reality check: near- vs. far-term applications
- Positron production, moderation, and trapping
- Solid parahydrogen (pH_2) as a positron moderator
- Slow electron transport experiments & simulations
- Positron moderation experiments
  - solid Ne moderator
  - solid pH_2 "moderator"
  - H_2O-doped solid Kr moderator
  - H_2O-ice coated Kr moderator
- Summary & Future Directions

Introduction - Positrons

- The positron (e^+) is the antiparticle to the electron (e^-).
- Positrons are stable, but annihilate with electrons to produce γ-rays.



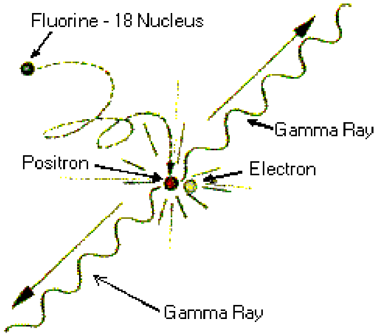
- "Positronium" (Ps) is a bound e^+/e^- pair; analogous to the H-atom (p^+/e^-).

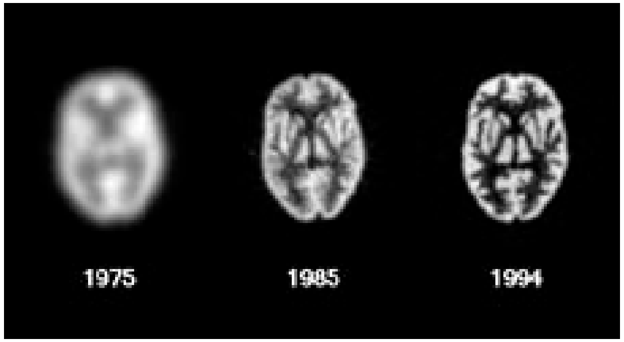


- Sum of γ-ray energies: $E \approx 2\, m_e c^2 \approx 1.02$ MeV; $\Rightarrow$ no photonuclear reactions or <u>secondary</u> radioactivity, but <u>lots</u> of energy/mass ($>10^{10}$ x chemical energies).
- However, bulk positron or positronium production and storage are problematic (understatement!).

Current Positron Applications

- 2-γ decay exploited in Positron Emission Tomography (PET) scanners.





- Positrons localize & annihilate preferentially at defects in materials. Basis for materials diagnostics:
  Doppler Broadening of Annihilation Radiation (DBAR) & Positron Annihilation Lifetime Spectroscopy (PALS).

Antimatter is real, and is utilized in hundreds of labs worldwide.

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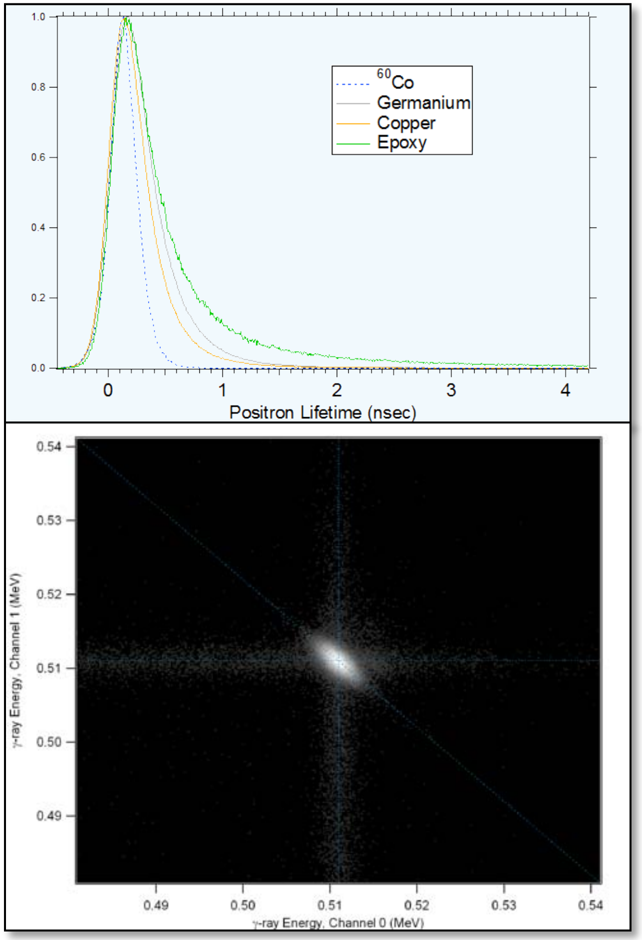
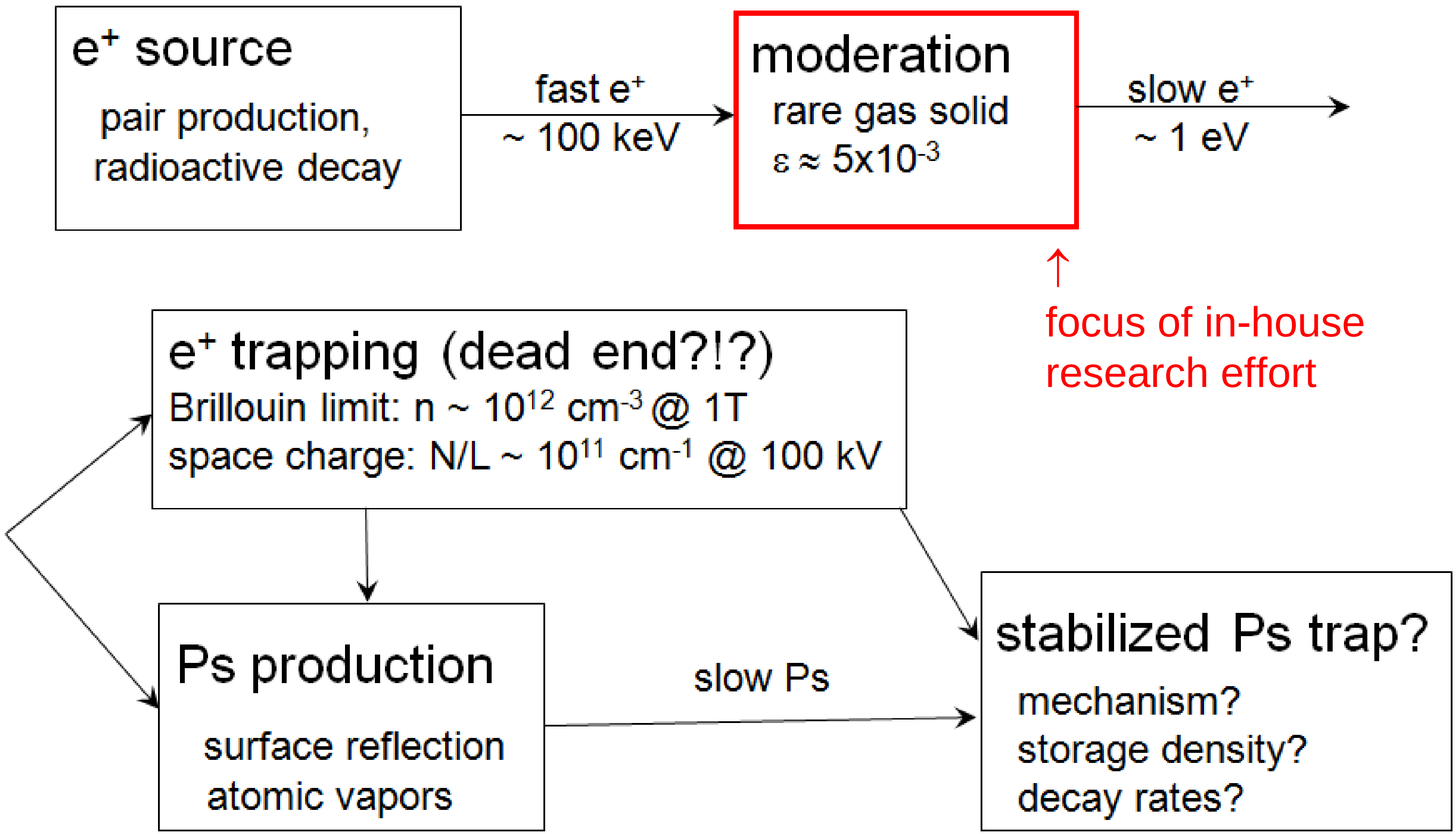
Energetic Materials Characterization

- Voids and defects play critical roles in energetic materials (EM).
- Few data available on nanoscale (sub-micron) defects.
- In-house NanoEnergetics effort underway to develop characterization tools to probe nanoscale defects in EM.
- Objective: measure defect size and concentration in EM and their relationship to processing and mechanical insults, and EM sensitivity.
- Accomplishments: PALS and 2D-DBAR instrumentation in use (home-built, pushing state-of-the-art).
- POC: Dr. C.M. Lindsay (ARFL/RWME).



Near-term payoffs from in-house positron investments.

Generic Positron Trapping Scheme



Moderation efficiency improvements are cumulative for slow e^+ applications.

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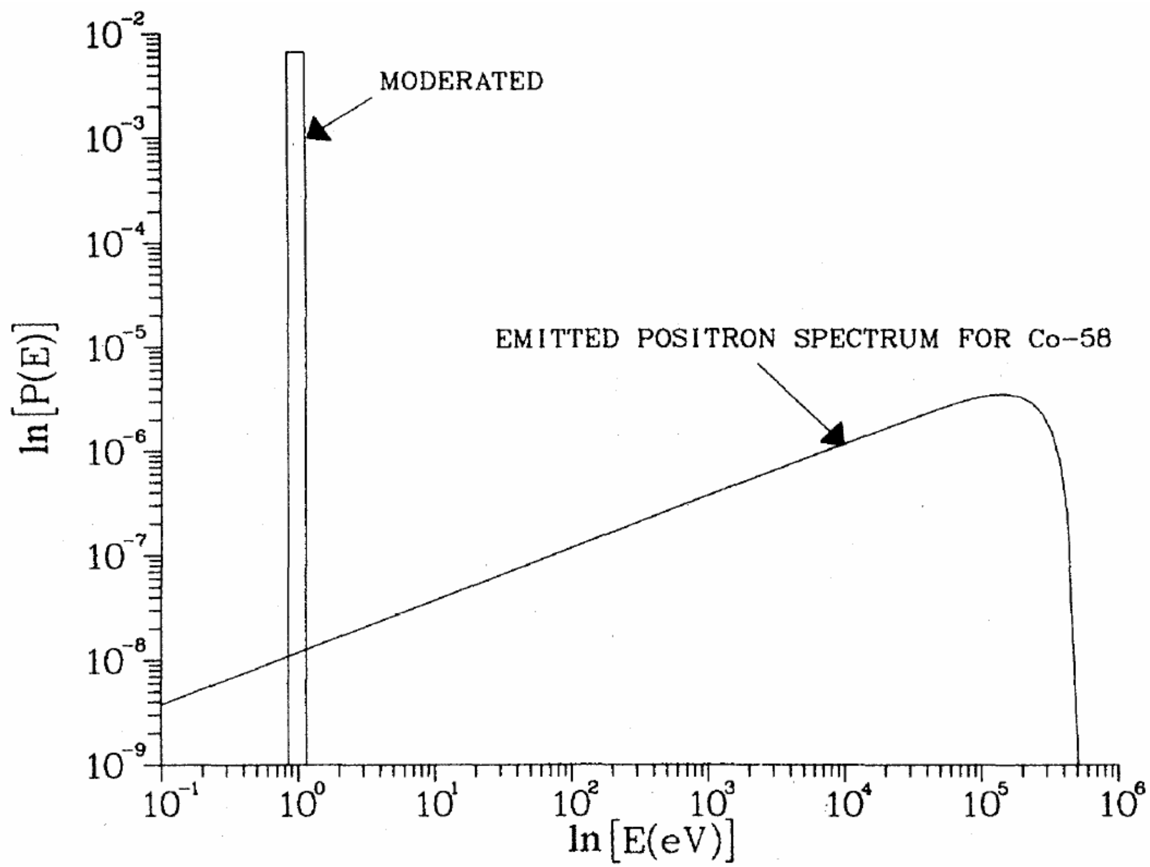
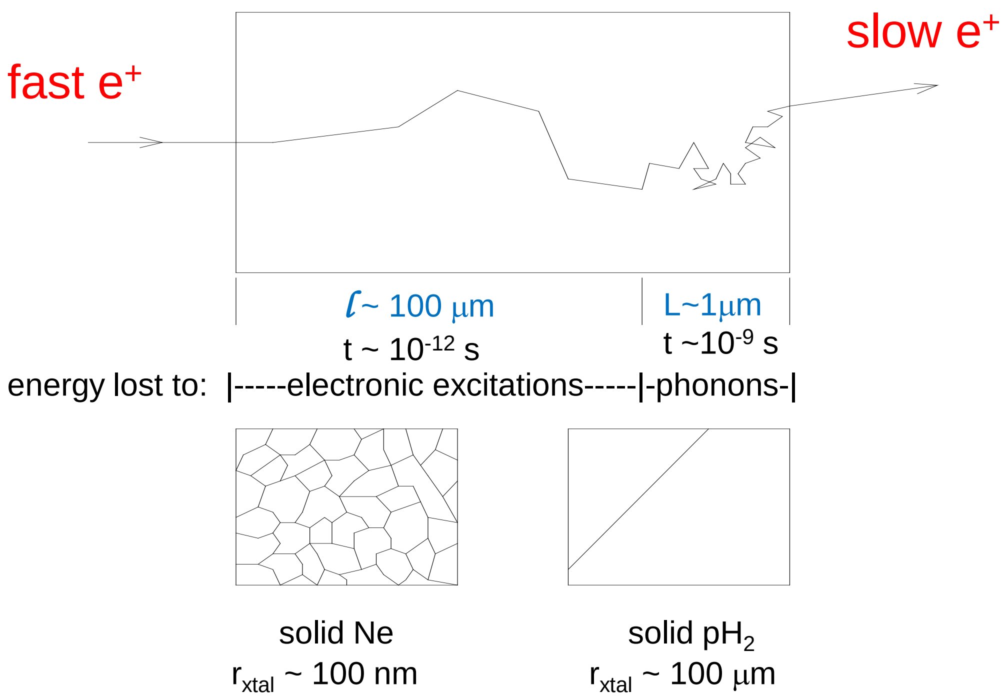
Moderation vs. Velocity Selection



P.J. Schultz and K.G. Lynn, Rev. Mod. Phys. **60**, 701 (1988).

Moderation converts fast positrons into slow positrons.

Positron Moderation Physics



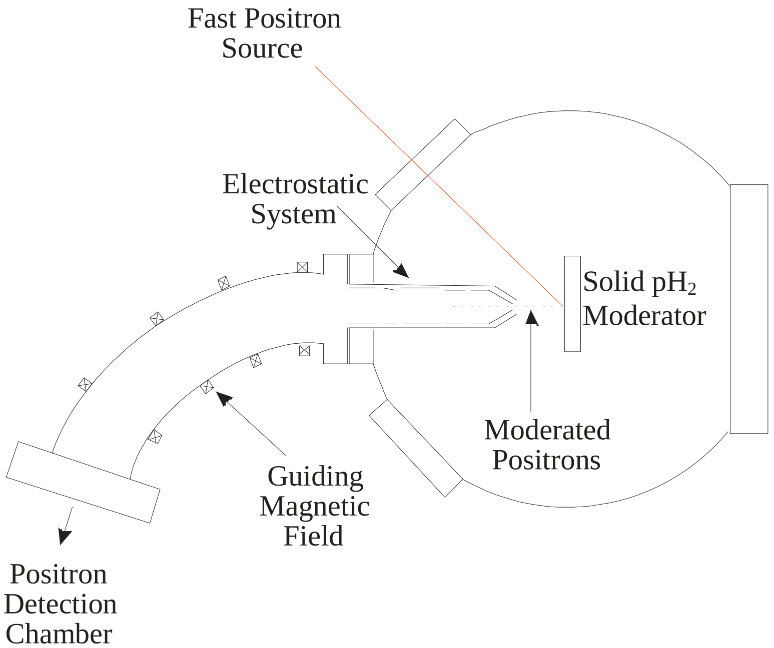
E.M. Gullikson and A.P. Mills, Jr., Phys. Rev. Lett. **57**, 376 (1986).

- Fast positrons decelerate rapidly in the first ~ picosecond by creation of electronic excitations.
- Slow positrons (below bandgap energy) can only create phonons and diffuse randomly for nanoseconds.
- Positrons which reach a moderator surface can be extracted & recovered.
- Slow positrons scatter, trap and annihilate at defects, becoming lost to the moderation process.
- <u>Conjecture</u>: higher quality RVD pH_2 solids should permit more slow positrons to emerge from deeper within the moderator.

Moderation efficiency increases as ~ L / ℓ.

Moderation Objectives & Approach

- Develop improved efficiency (ε) positron moderators based on cryogenic (T ≈ 2 K) RVD pH_2 solids
  goal: 10x improvement over the state of the art (Ne), yielding ε ≈ 5 %.
- Determine preferred pH_2 moderator structures and moderator failure modes by *in-situ* spectroscopic characterization.
- Moderate fast positrons from ~ 1 mCi ^{22}Na source in reflection geometry.
- Separate slow from fast positrons using electrostatic and magnetic guiding fields around bend into annihilation chamber.
- Slow positron detection: charged particle and annihilation γ-radiation.

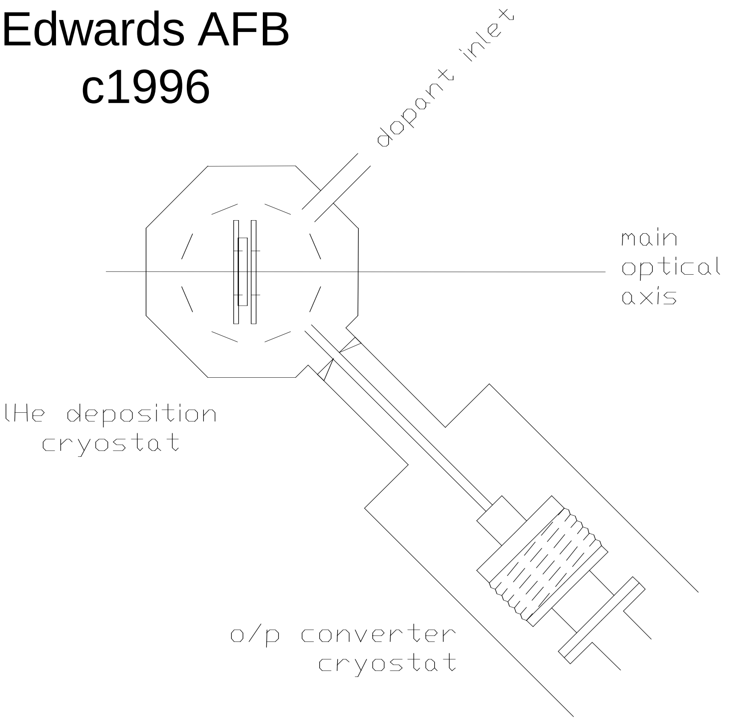


Notional Experimental Diagram

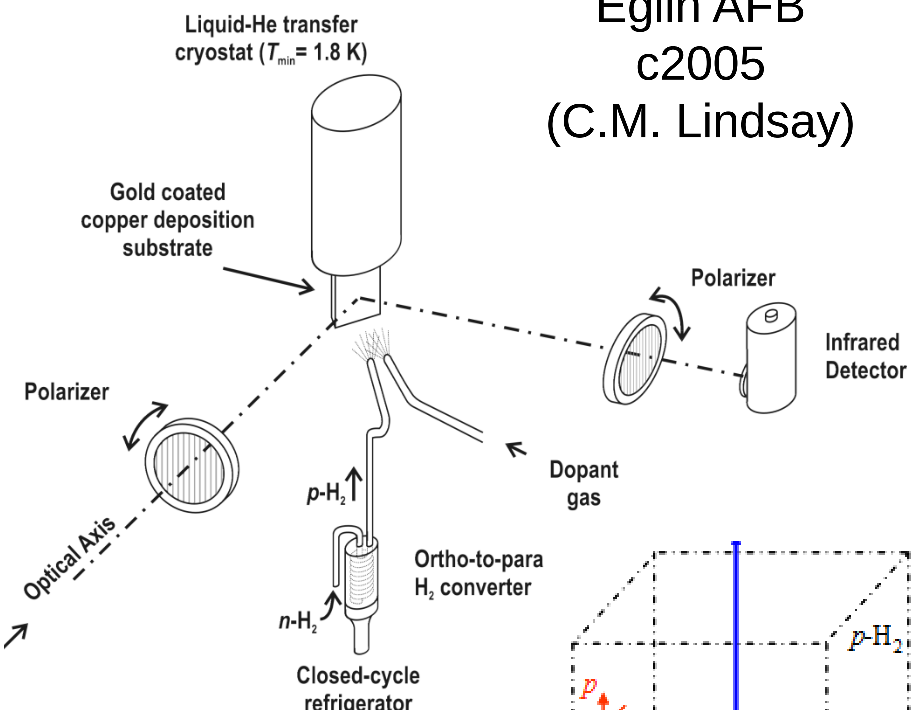
C.D. Molek, "Moderation of Fast Positrons Using Solid Parahydrogen," NRC Research Proposal (2007).

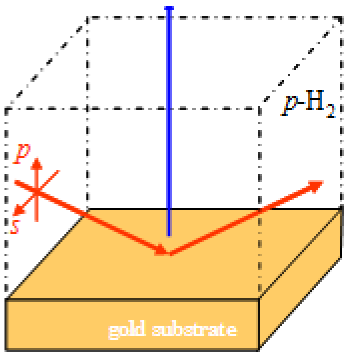
Rapid Vapor Deposition of Solid pH_2

Edwards AFB
c1996



Eglin AFB
c2005
(C.M. Lindsay)

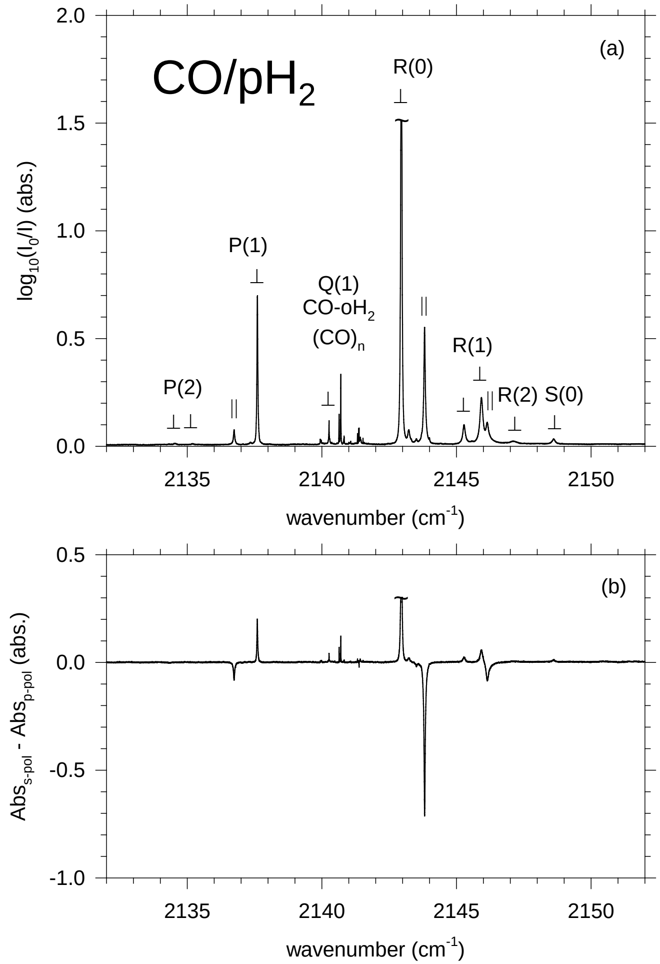


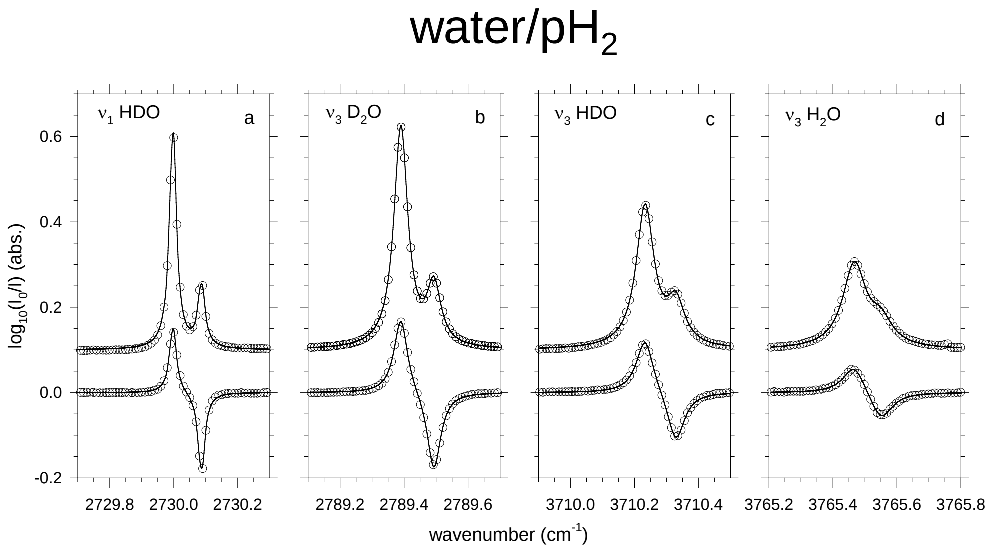


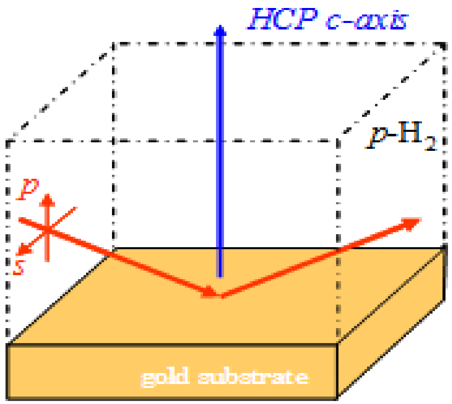
M.E. Fajardo and S. Tam, J. Chem. Phys. **108**, 4237 (1998).
S. Tam and M.E. Fajardo, Rev. Sci. Instrum. **70**, 1926 (1999).
M.E. Fajardo and C.M. Lindsay, J. Chem. Phys. **128**, 014505 (2008).
M.E. Fajardo, C.M. Lindsay, and T. Momose, J. Chem. Phys. **130**, 244508 (2009).

Building upon solid pH_2 expertise developed during 1990's "HEDM" program.

Polarized IR Absorption Spectra





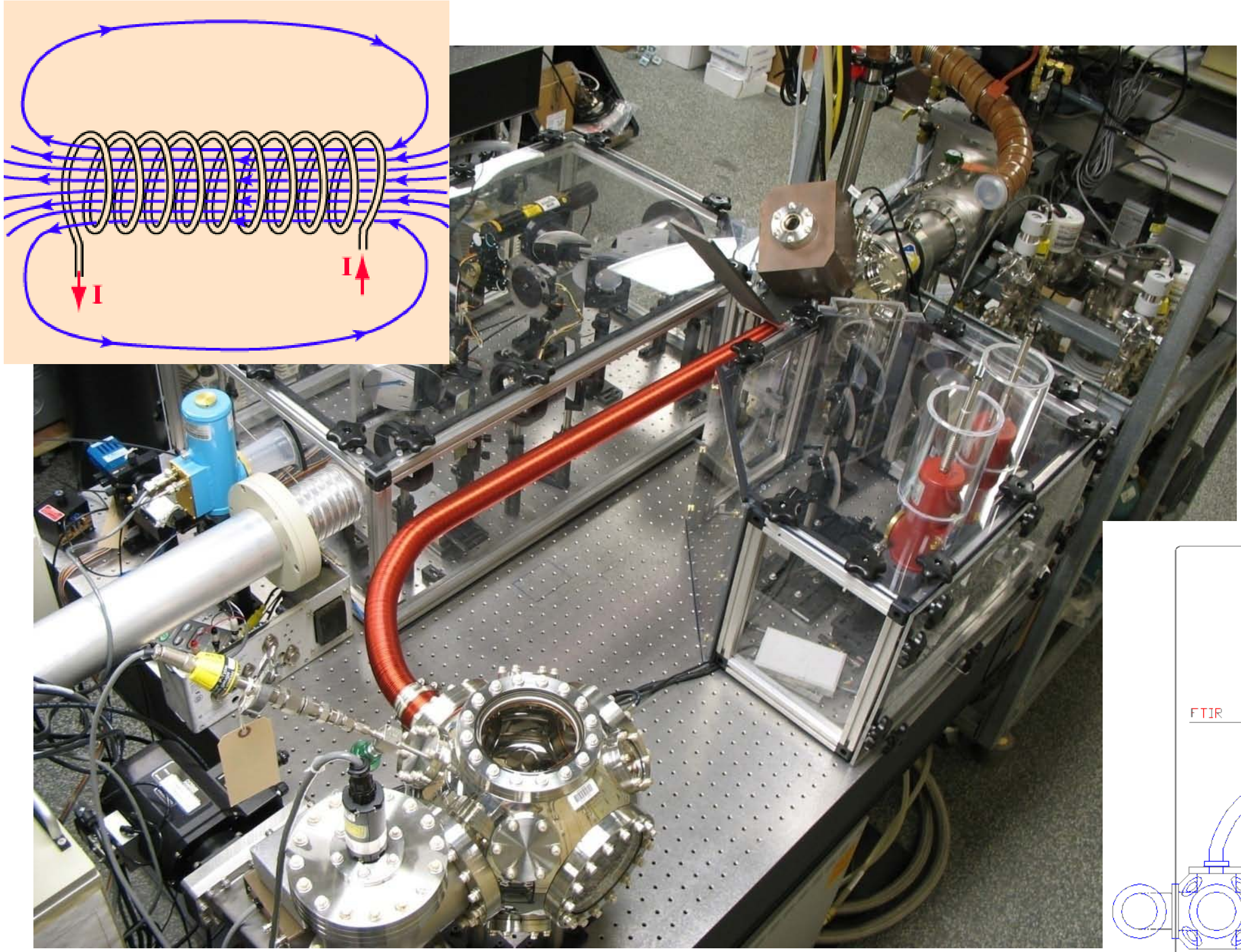


Polarization dependence as predicted by Crystal Field Theory modeling.

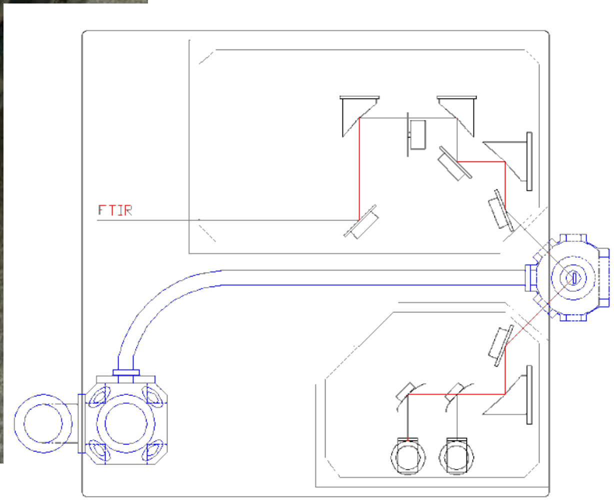
M.E. Fajardo and C.M. Lindsay, J. Chem. Phys. **128**, 014505 (2008).
M.E. Fajardo, C.M. Lindsay, and T. Momose, J. Chem. Phys. **130**, 244508 (2009).

Annealed hcp c-axes <u>all</u> parallel to substrate surface normal! Single crystal?

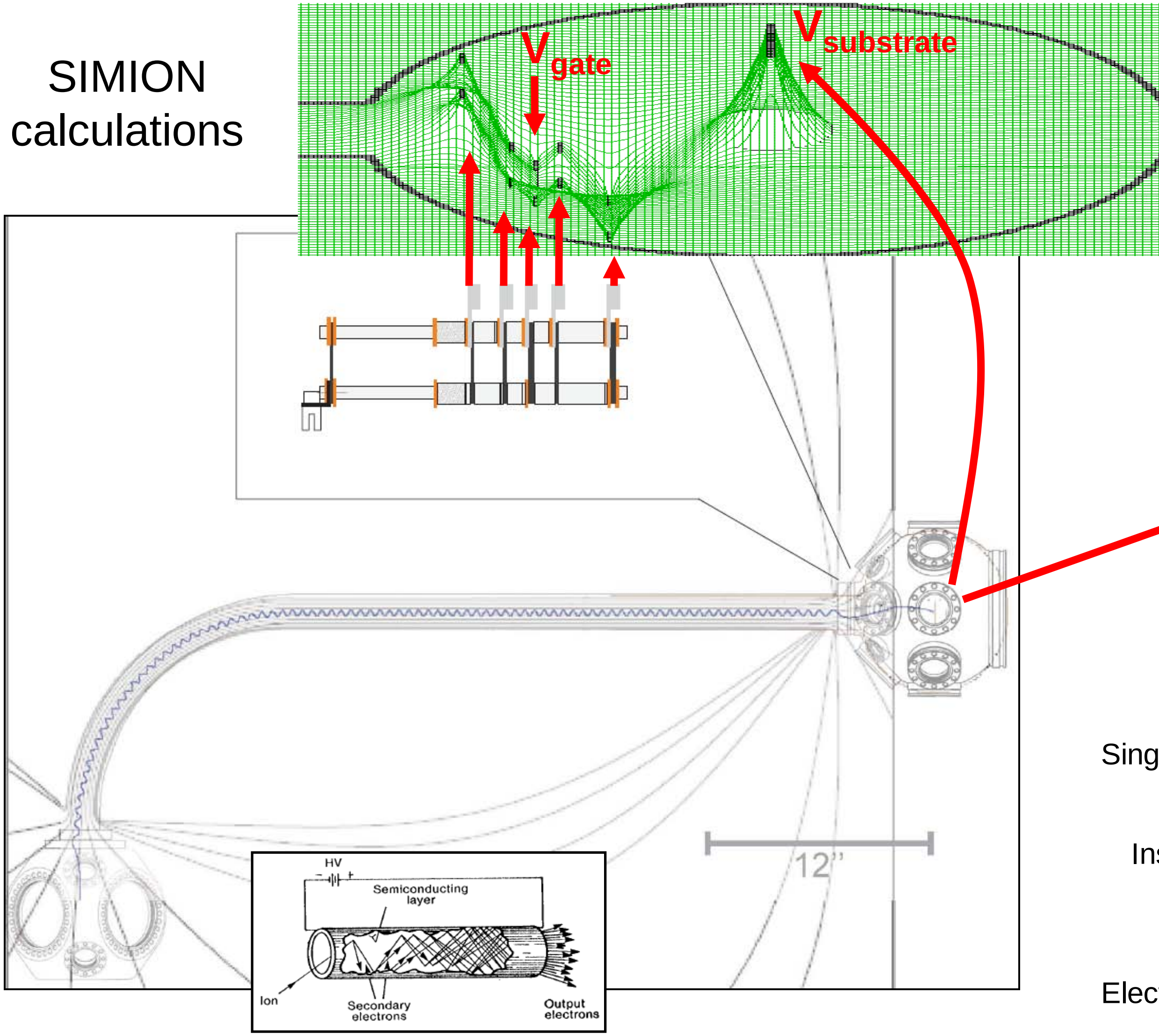
Preliminary Electron Transport Expt.

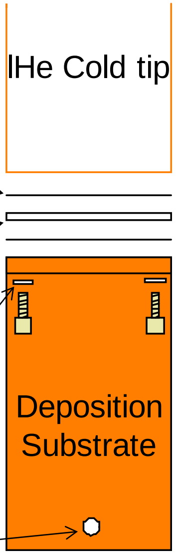


Shown configured for initial slow electron transport experiments through bent solenoid using hot-filament electron source and Faraday cup detector.



Ion Optics & Deposition Substrate





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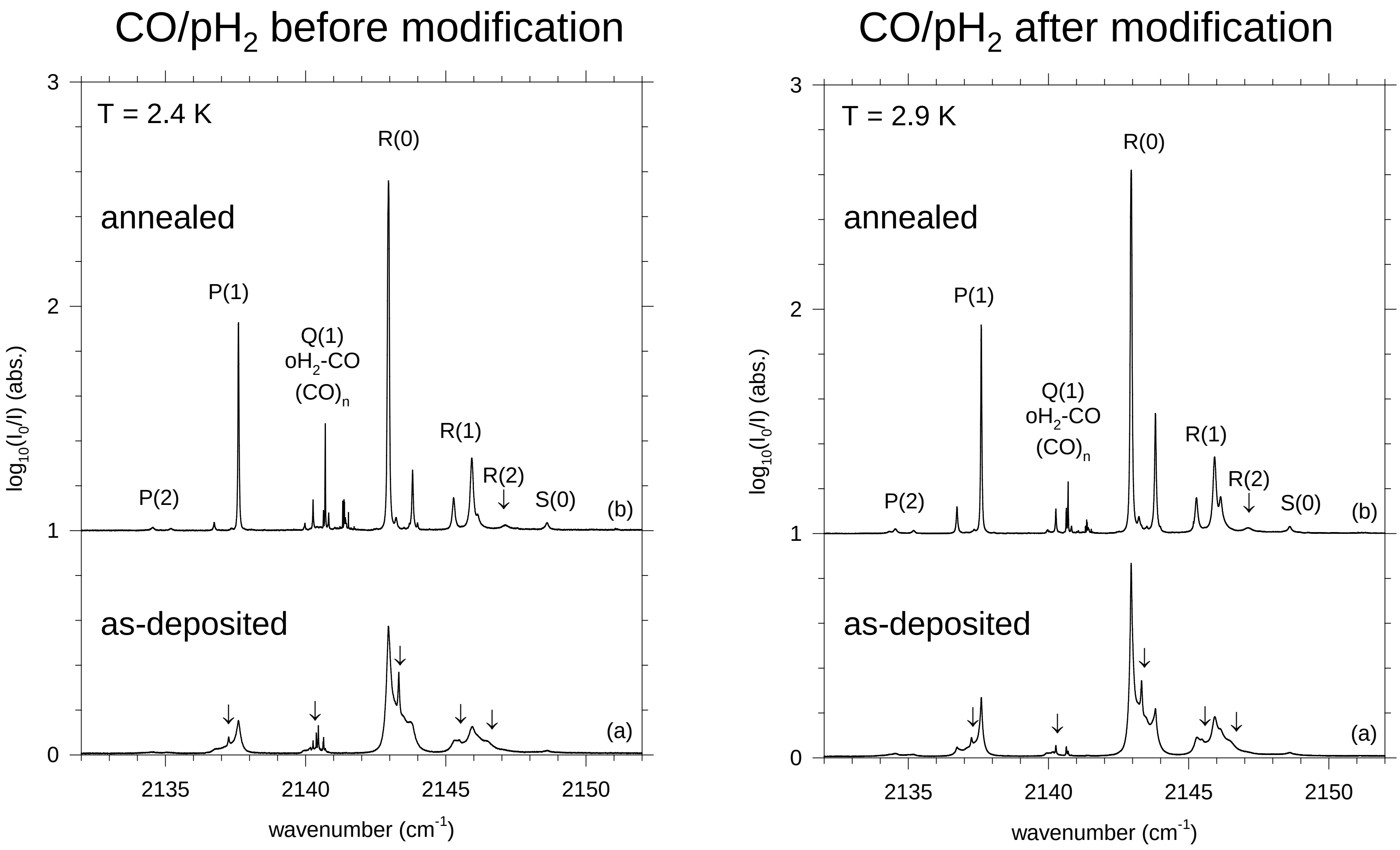
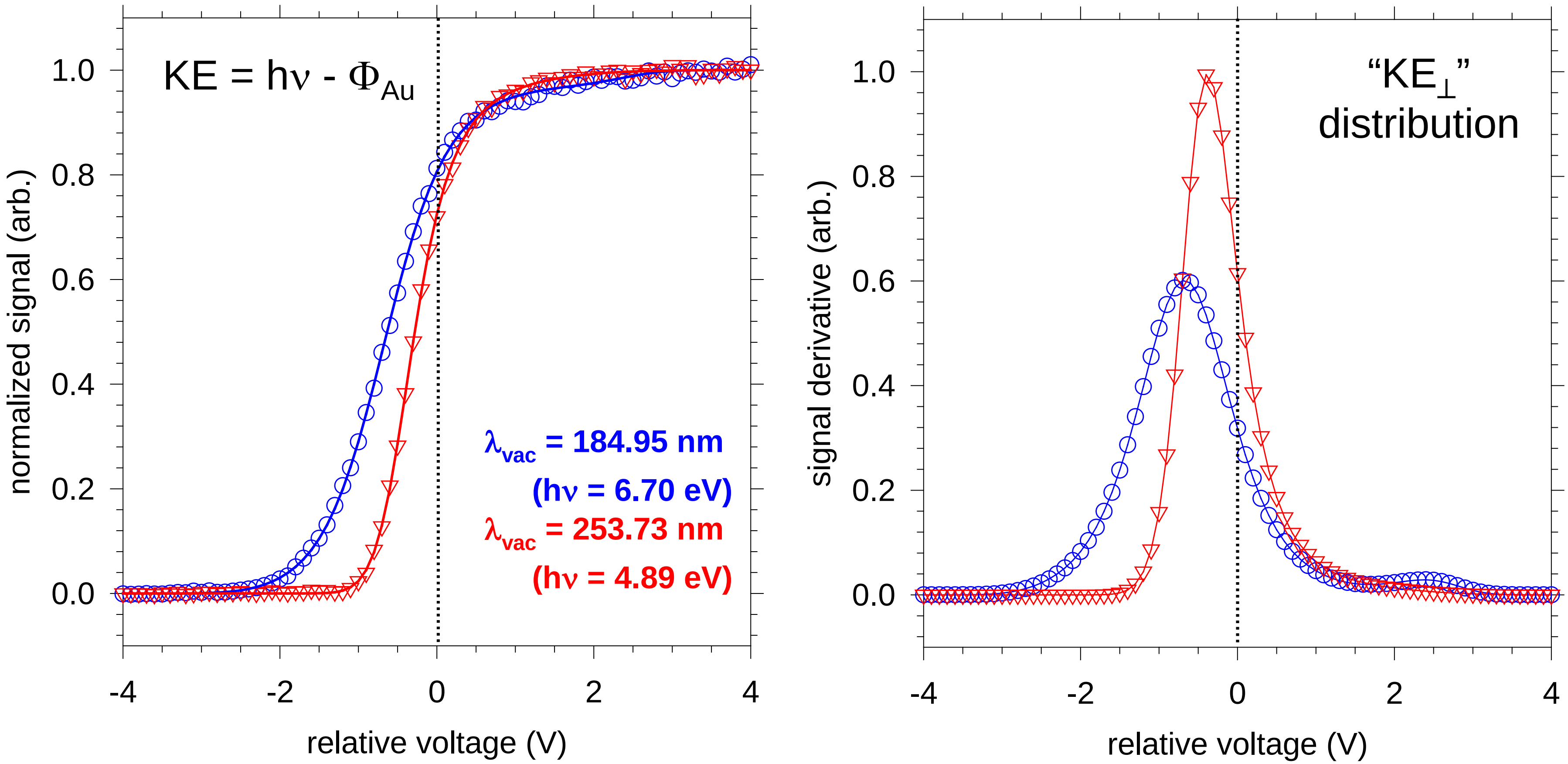
Isolated Deposition Substrate



Only minor changes to spectra from ≈ 0.5 K higher substrate temperature.

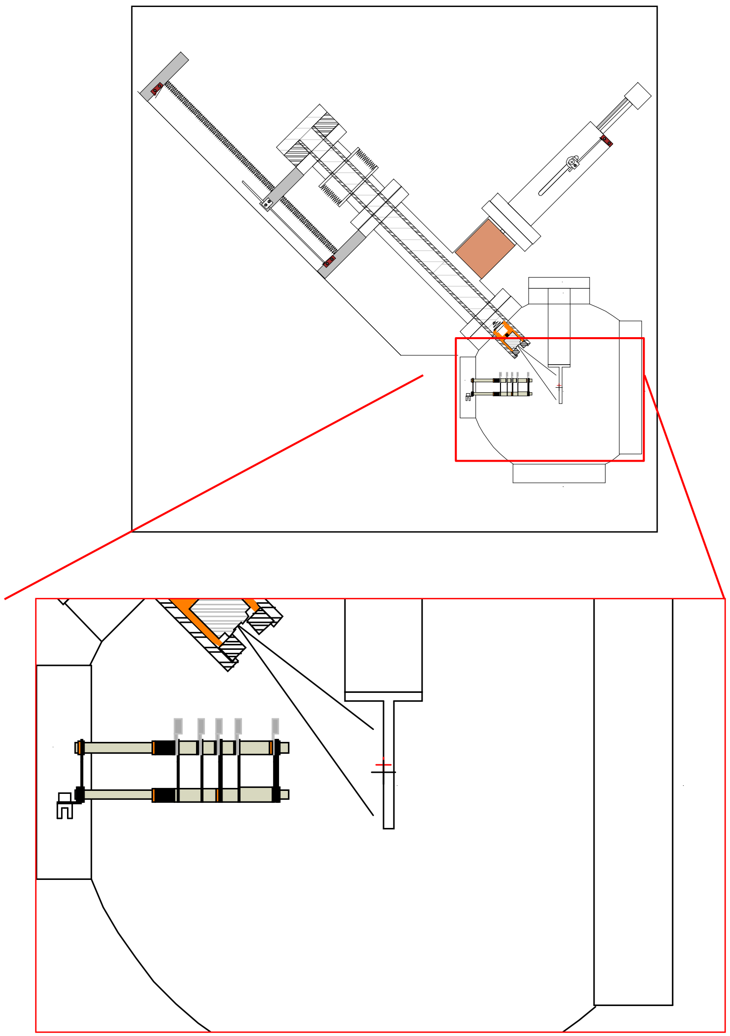
Photoelectron Transport Expts.

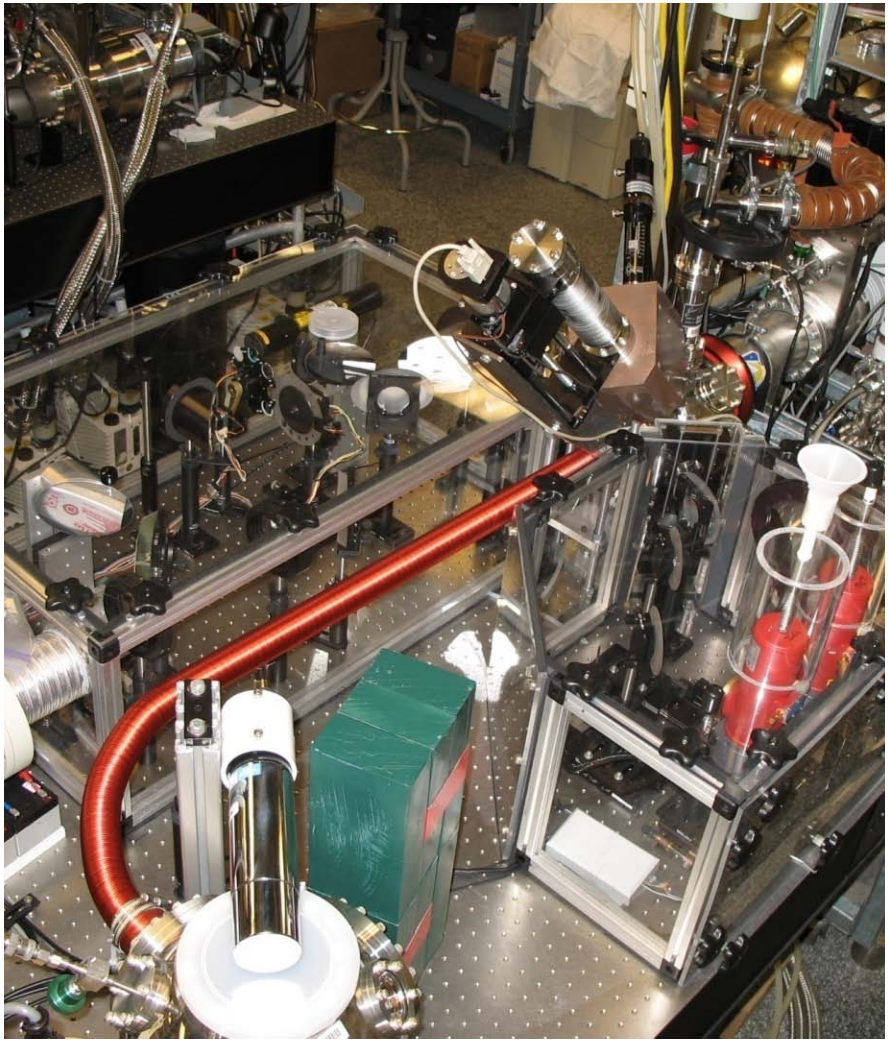
Hg/Ar pen lamp + UV bandpass filters + pinhole apertures



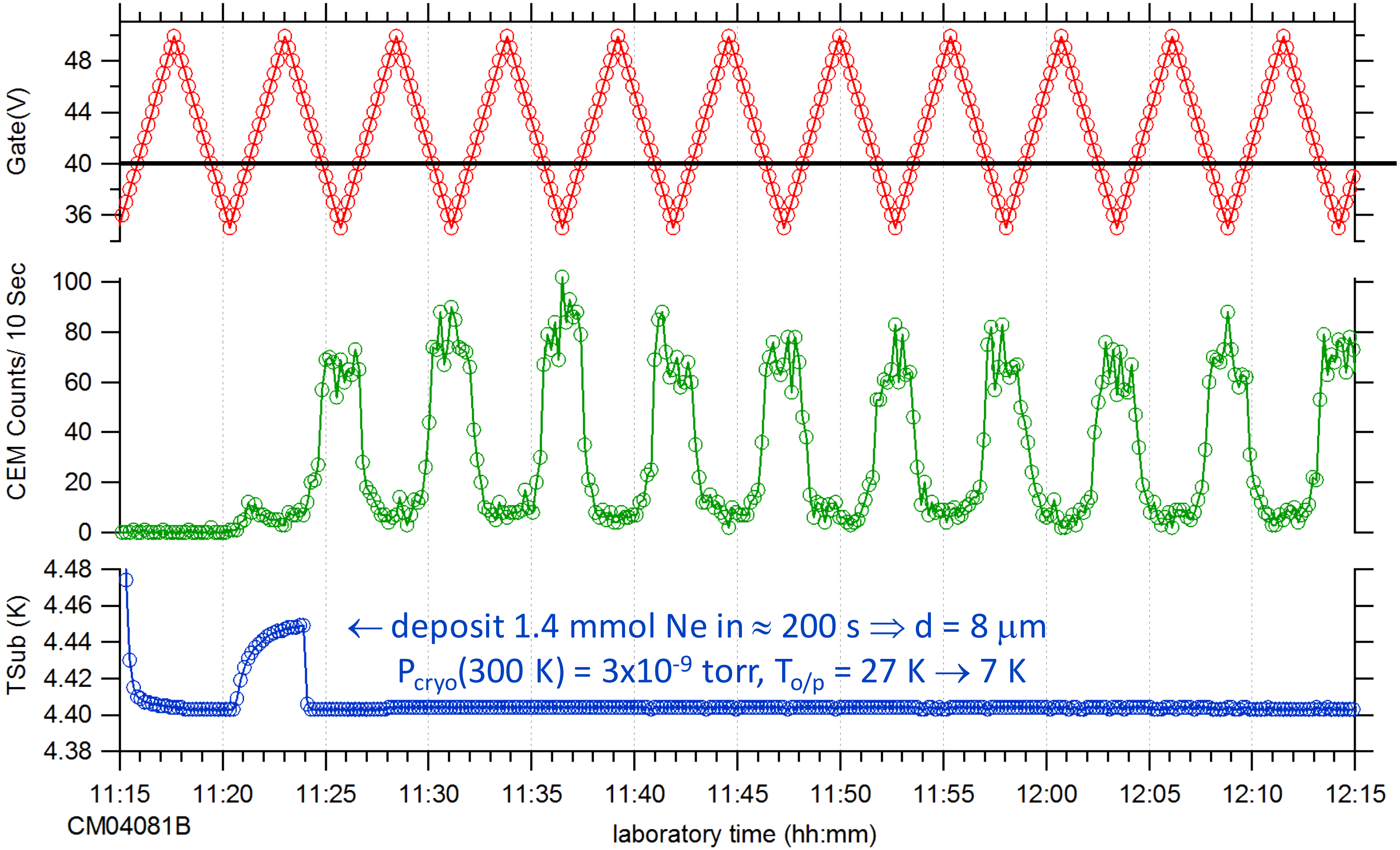
Electron throughput and energy resolution acceptable for e^+ moderation expts.

^{22}Na Positron Source – 1 mCi

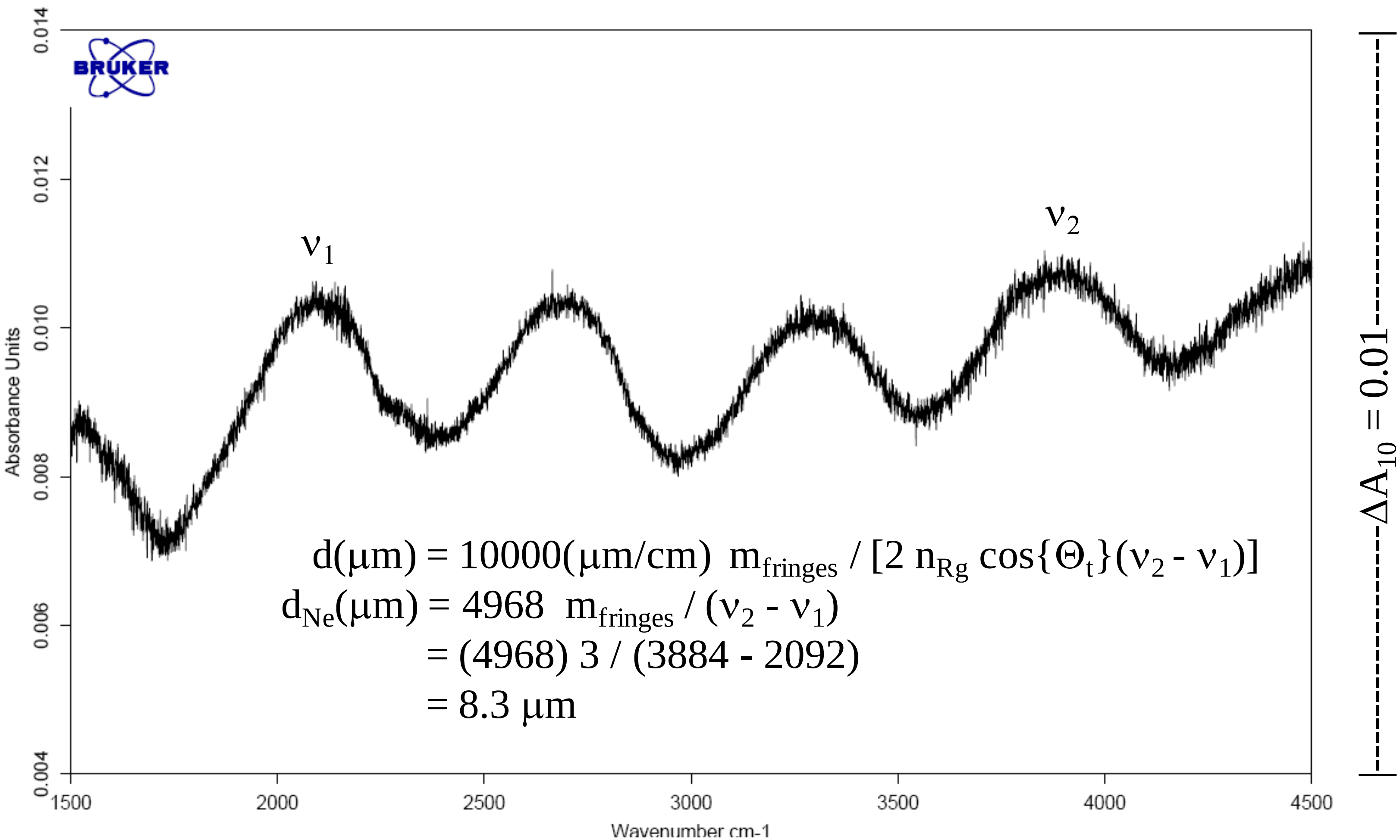




Ne Moderator Deposition



Ne Moderator Thickness



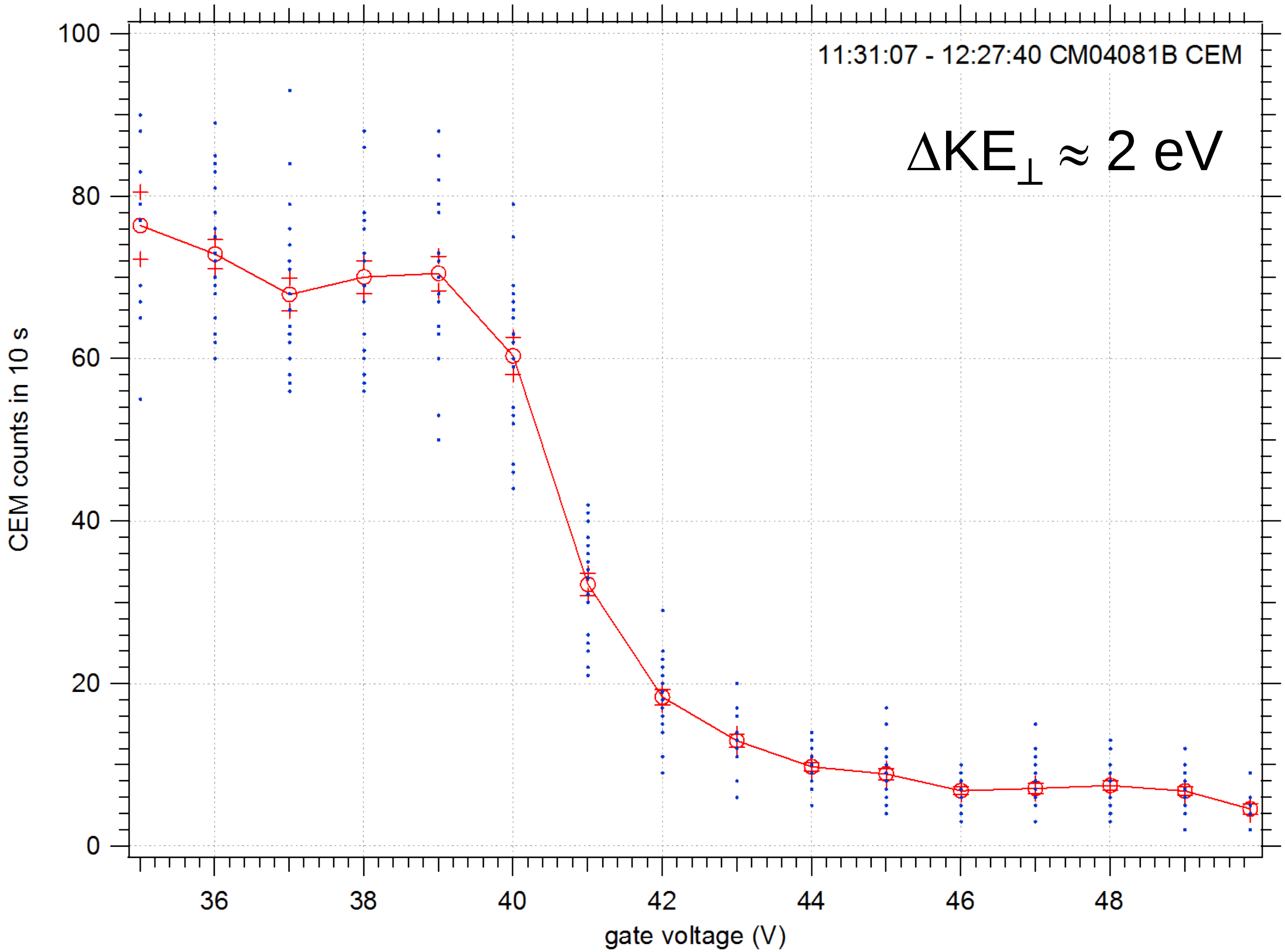
$$d(\mu m) = 10000(\mu m/cm)\ m_{fringes} / [2\, n_{Rg} \cos\{\Theta_t\}(\nu_2 - \nu_1)]$$

$$d_{Ne}(\mu m) = 4968\ m_{fringes} / (\nu_2 - \nu_1)$$

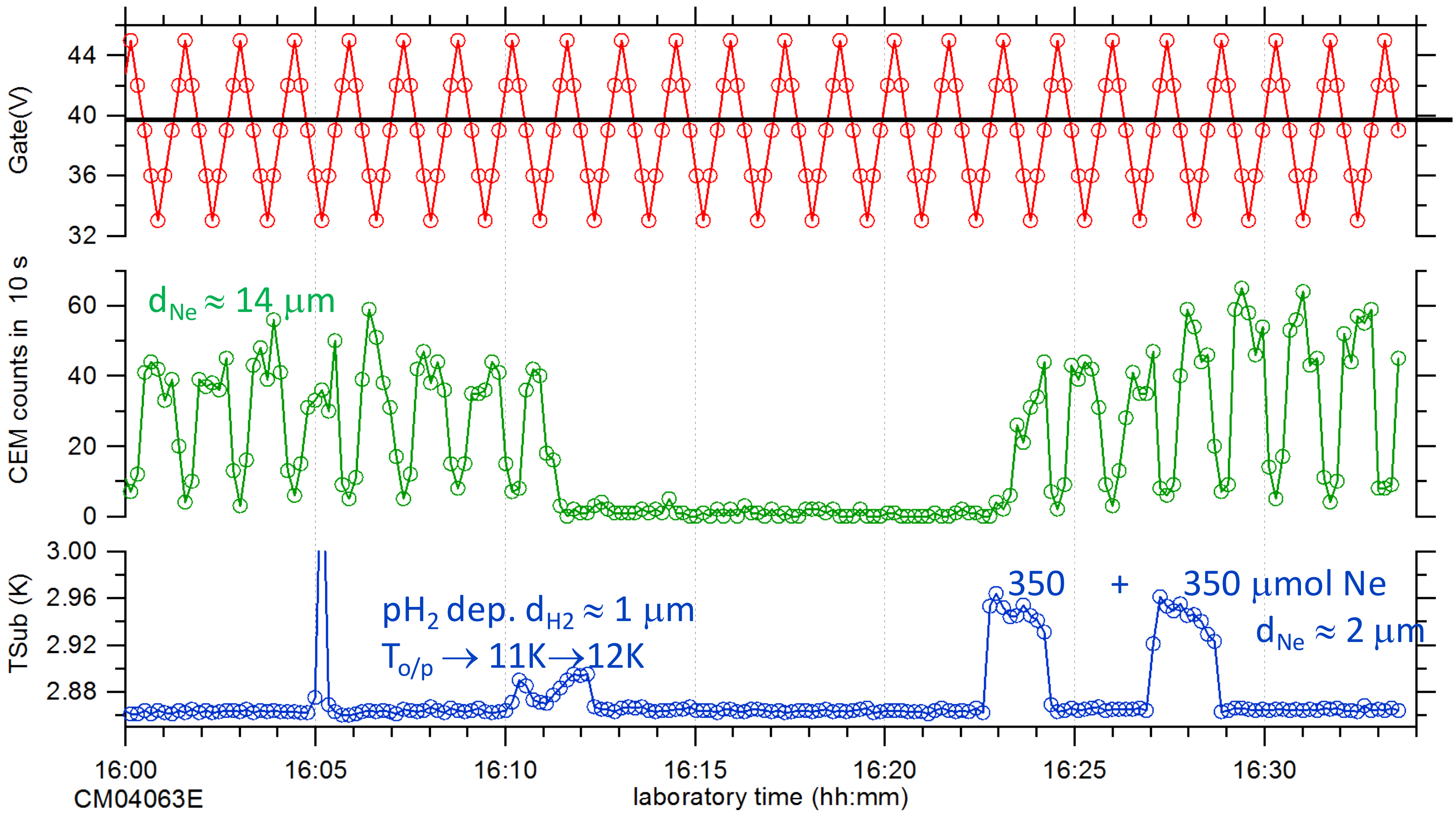
$$= (4968)\ 3 / (3884 - 2092)$$

$$= 8.3\ \mu m$$

Ne Moderator Performance

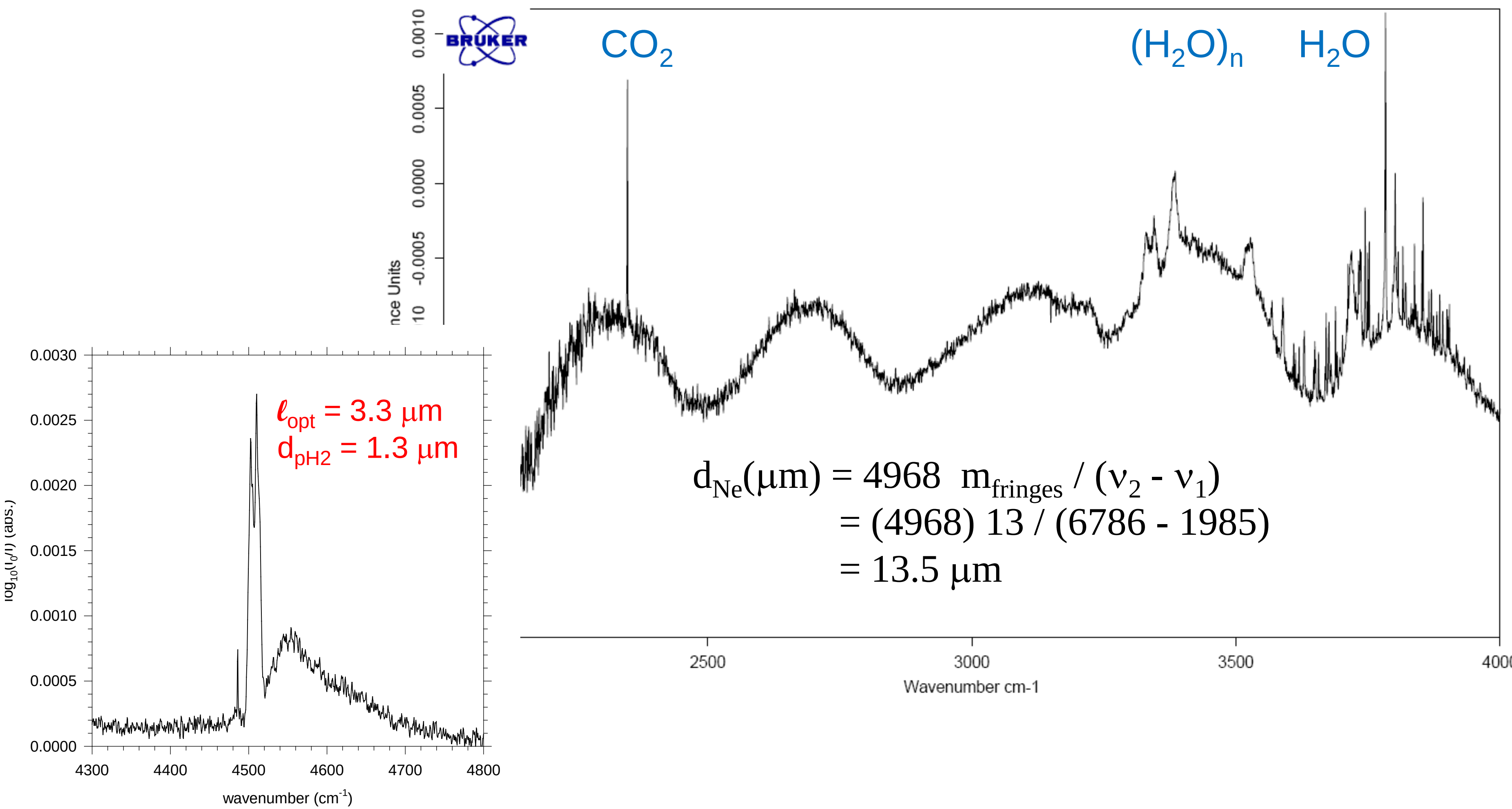


500 ppm oH_2/pH_2 Overcoat



Solid pH_2 appears to be a very poor positron moderator!

Dirty Ne Moderator + pH_2 Overcoat



$$d_{Ne}(\mu m) = 4968 \; m_{fringes} / (\nu_2 - \nu_1)$$
$$= (4968)\, 13 / (6786 - 1985)$$
$$= 13.5 \; \mu m$$

Ne deposited from dopant manifold. $P_{cryo}(300K) = 1.1\text{x}10^{-8}$ torr.

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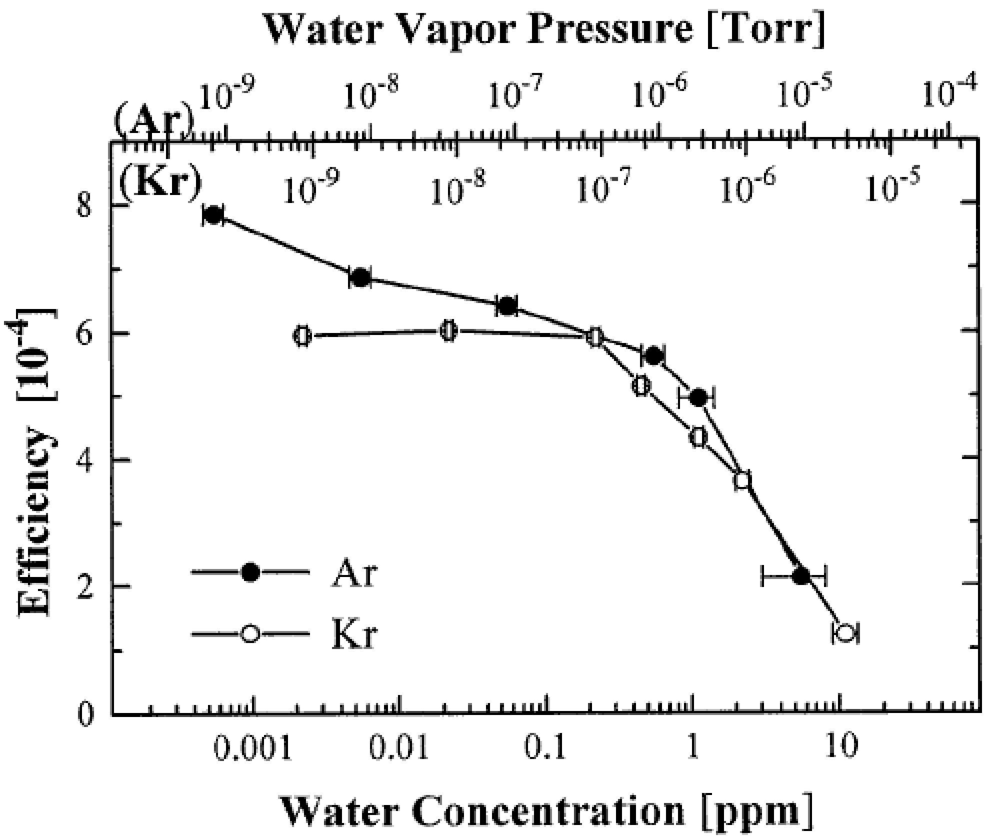
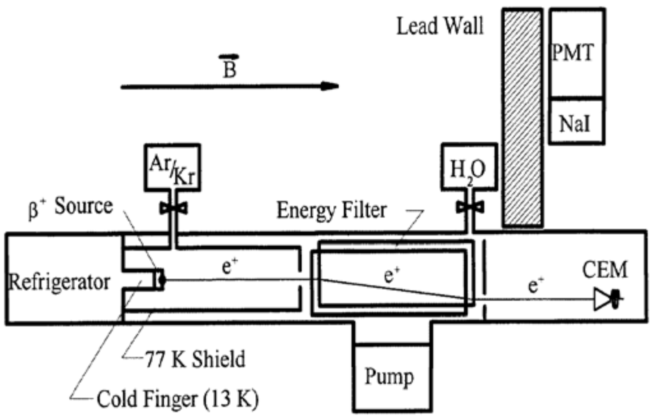
Effects of H_2O Impurities

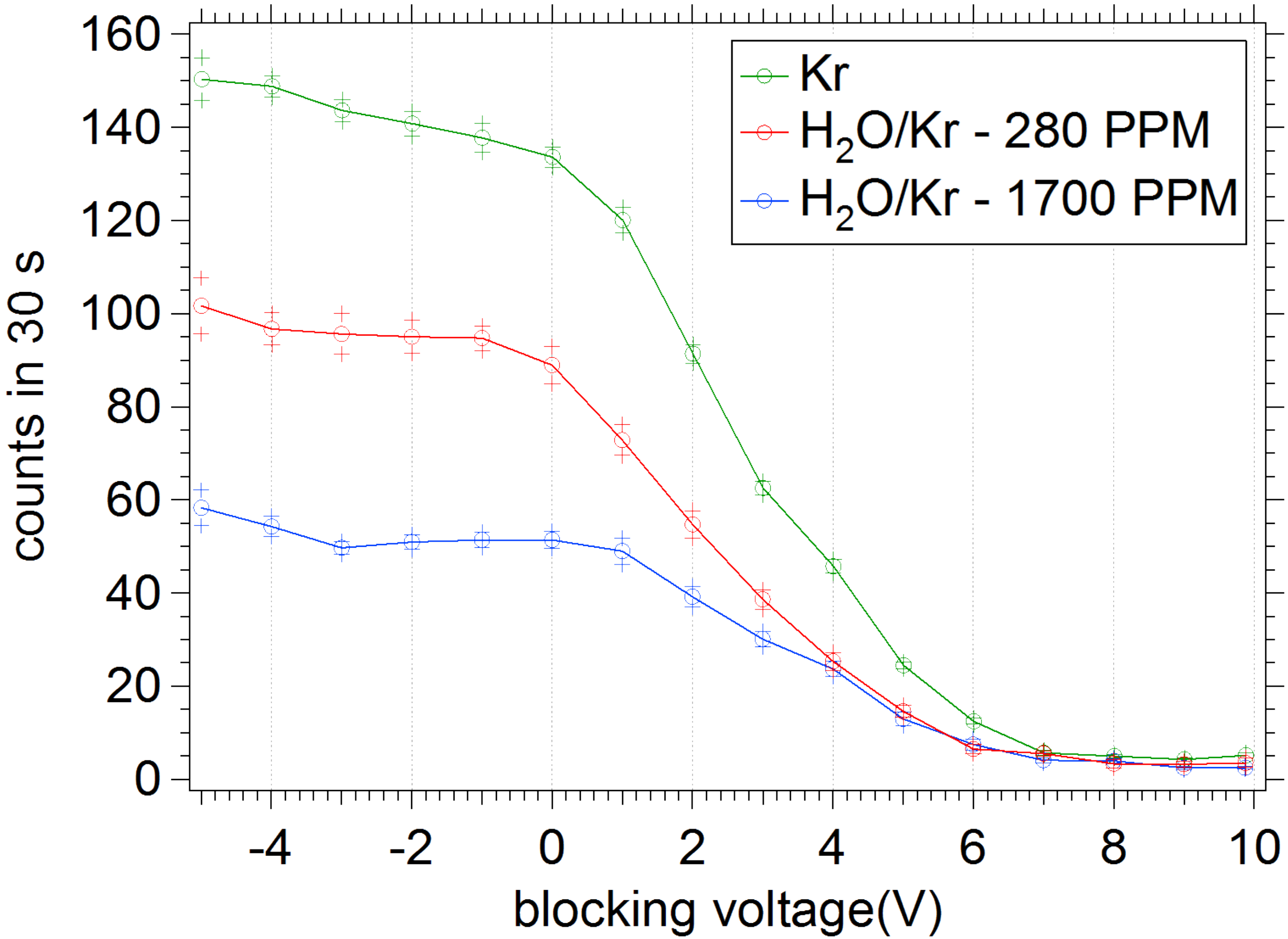




H_2O concentrations estimated from H_2O partial pressures in deposition chamber.

M.P. Petkov, K.G. Lynn, and L.O. Roellig, J. Phys. Cond. Mat. **8**, L611 (1996).

H_2O/Kr Moderators



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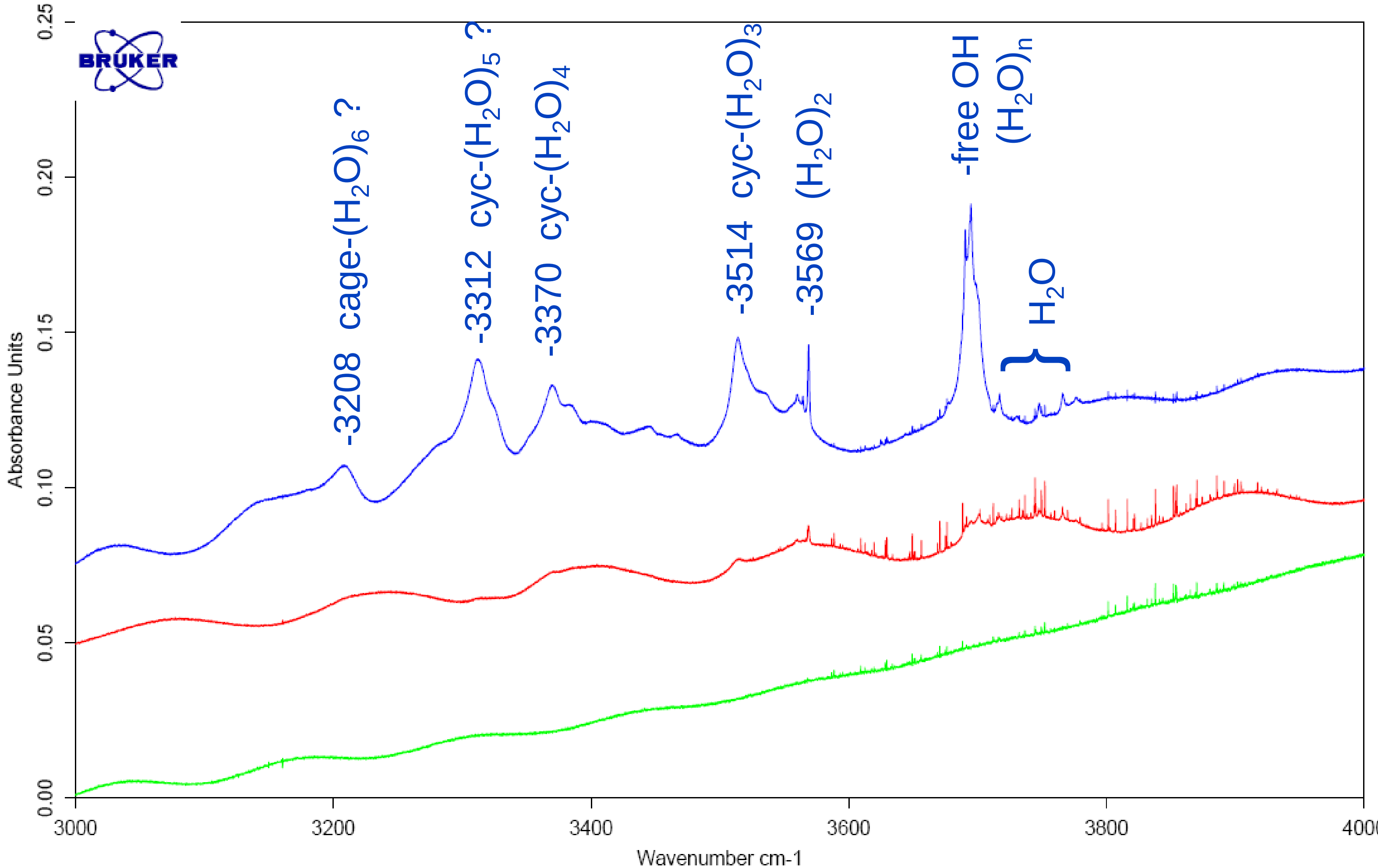
H_2O/Kr Moderators



nominal H_2O conc. & thickness

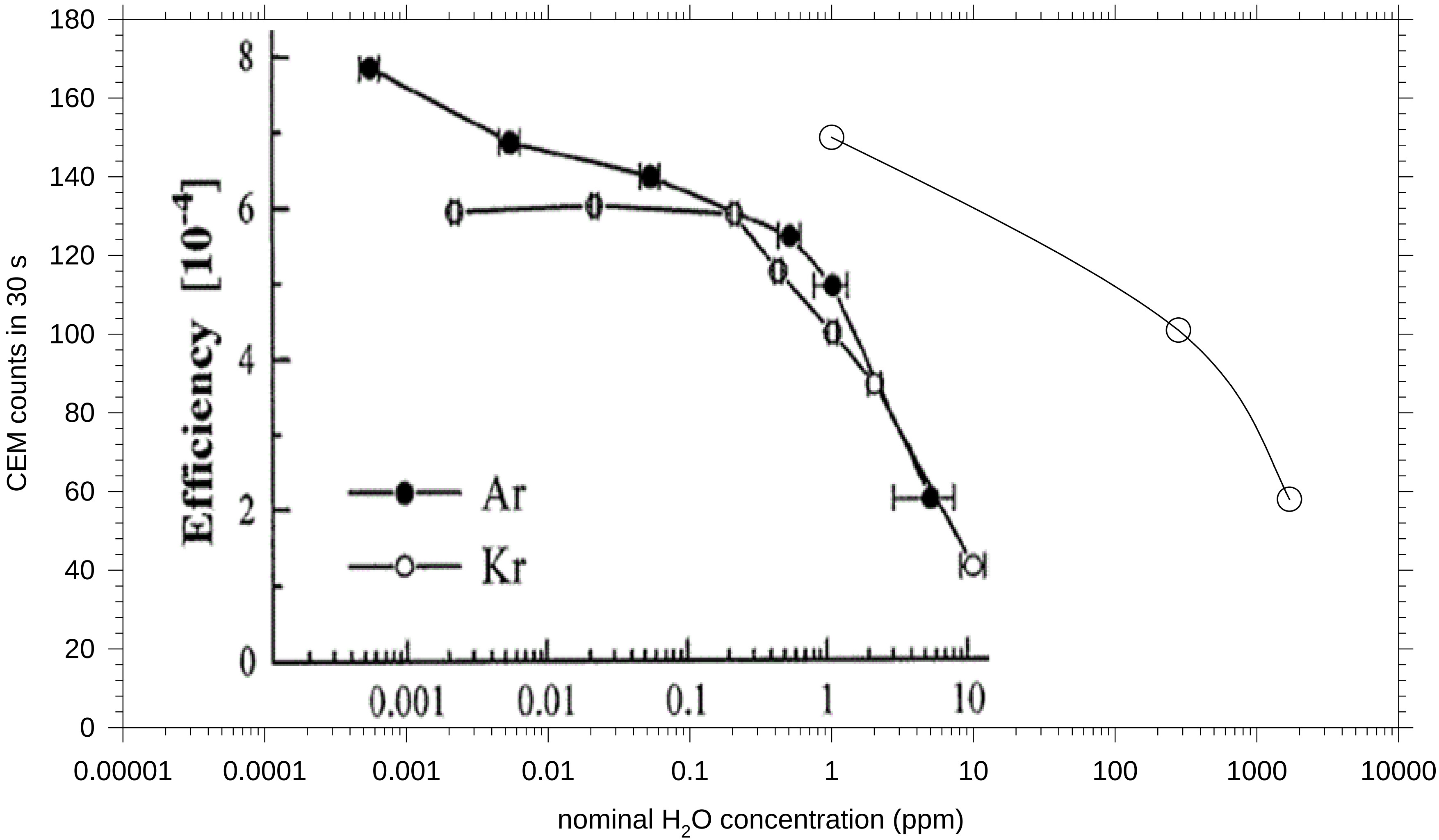
1700 ppm
d = 33 μm

280 ppm
d = 25 μm

~1(<20) ppm
d = 31 μm

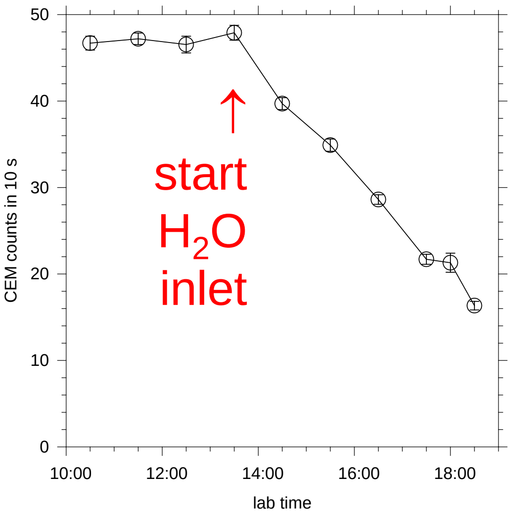
$(H_2O)_n$/Kr assignments – A. Engdahl and B. Nelander, J. Mol. Struct. **193**, 101 (1989).

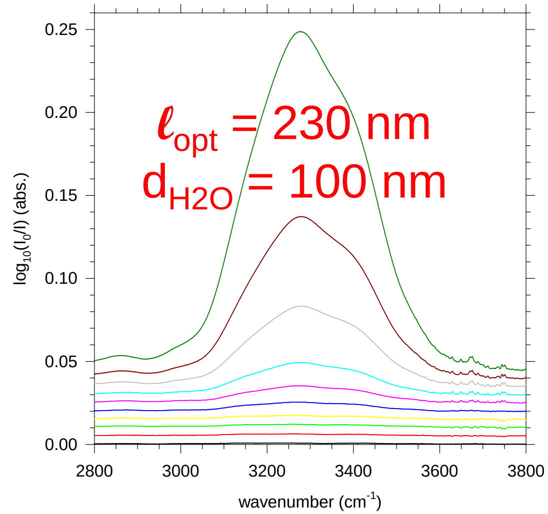
H_2O/Kr Moderators

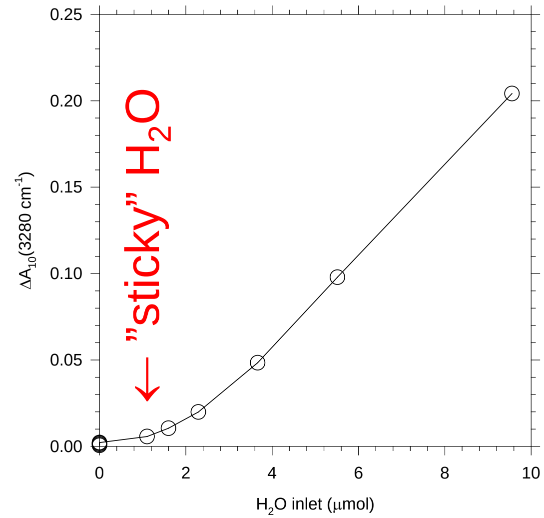


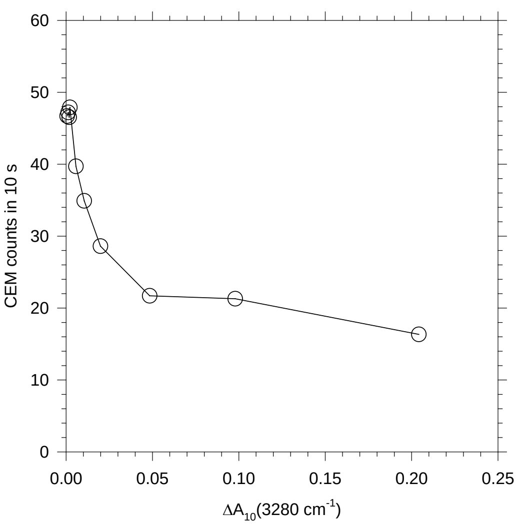
H_2O concentration estimates differ by two orders of magnitude.

H_2O Ice Overcoat on Kr Moderator









FTIR spectrum consistent with amorphous ice, as expected for vapor deposition with $T_{sub} = 30$ K.
[W. Hagen, A.G.G.M. Tielens, and J.M. Greenberg, CP **56**, 367 (1981).]

See initial rapid drop in slow e^+ yield, as expected.

Plateau in slow e^+ signal for thick ice <u>not</u> expected;
$\Rightarrow$ porous ice, possible experimental artifact (?!).

Summary & Future Directions

- Demonstrated an apparatus for combined positron moderation + matrix isolation spectroscopy; permits testing of theories proposed to explain observed moderator efficiency reductions upon extended operation.

- Unfortunately, ~ 500 ppm oH_2/pH_2 cryogenic solids are poor e^+ moderators; $\varepsilon_{pH2} < 0.01\ \varepsilon_{Ne}$.

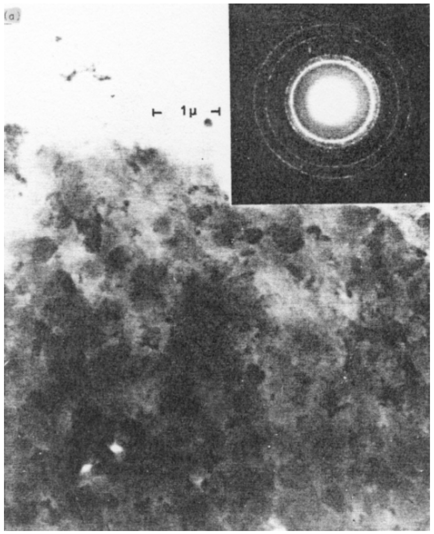
- Previous studies of the role of water impurities in Rg moderators may have seriously underestimated trapped H_2O concentrations.

- Slow positrons can emerge from Kr moderator coated with $\approx$ 100 nm amorphous water ice.

- One last set of experiments planned for the end of FY11. Will repair ortho/para hydrogen converter to bring residual oH_2 concentrations below 100 ppm.

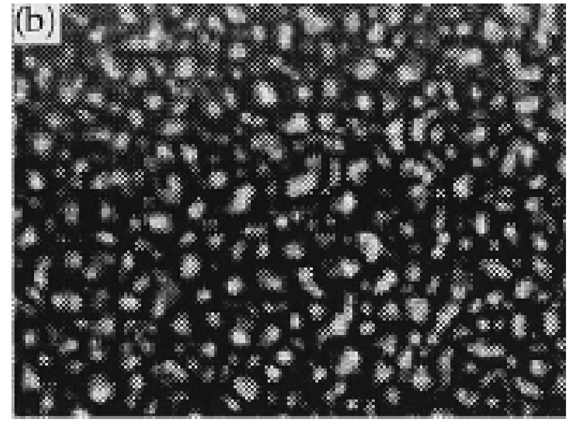
Backup Slides

Solid Ne vs. Solid pH_2



Electron micrograph and diffraction pattern of vapor deposited solid Ne showing sub-micron scale defects.

J.A. Venables and B.L. Smith, "Crystal Growth and Crystal Defects," in Rare Gas Solids Vol. II, edited by M.L. Klein and J.A. Venables (Academic Press, London, 1977).



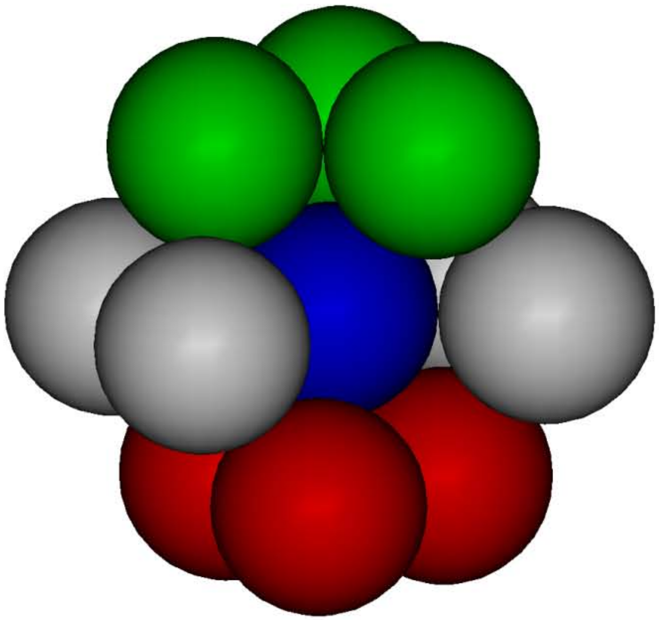
|------------ 1.7 mm ------------|

Typical crystallite size in vapor deposited pH_2 is ~ 100 μm, or ~1000x larger than for vapor deposited Ne.

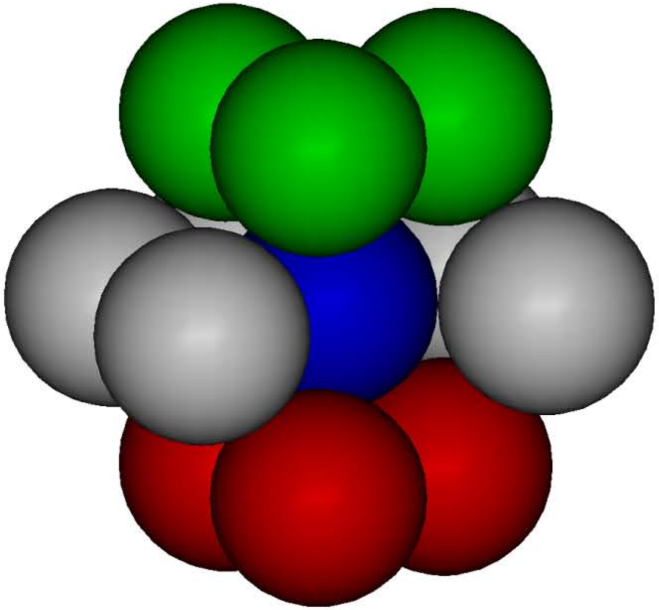
G.W. Collins, W.G. Unites, E.R. Mapoles, and T.P. Bernat, Phys. Rev. B **53**, 102 (1996).

Also: solid pH_2 is a special material – a "quantum solid."

fcc & hcp Structures



“face-centered cubic”
fcc $\Rightarrow$ O_h site symmetry,
nearly isotropic

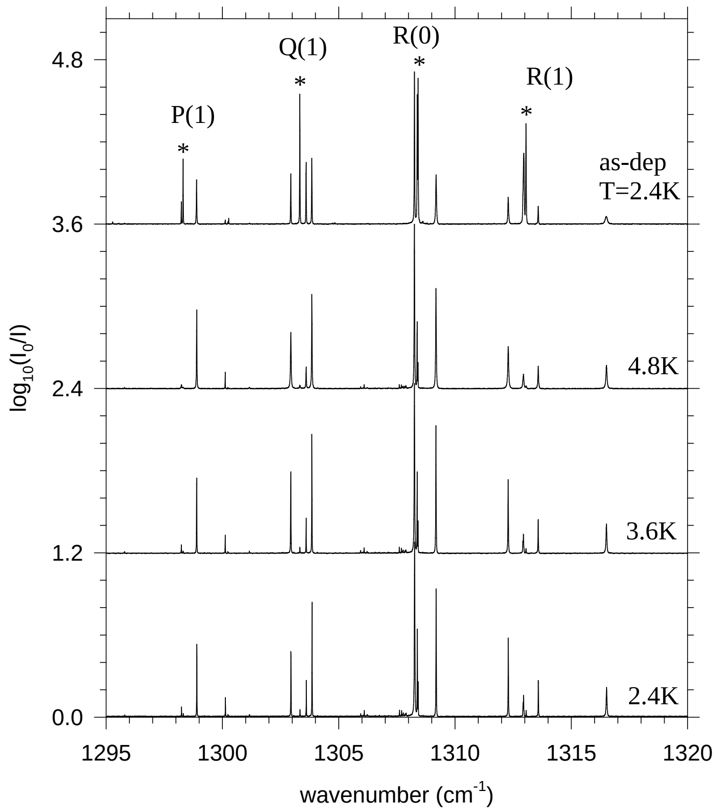


“hexagonal close-packed”
hcp $\Rightarrow$ D_{3h} site symmetry,
unique c-axis

Use dopant molecules in solid pH_2 as probes to determine crystal structure.

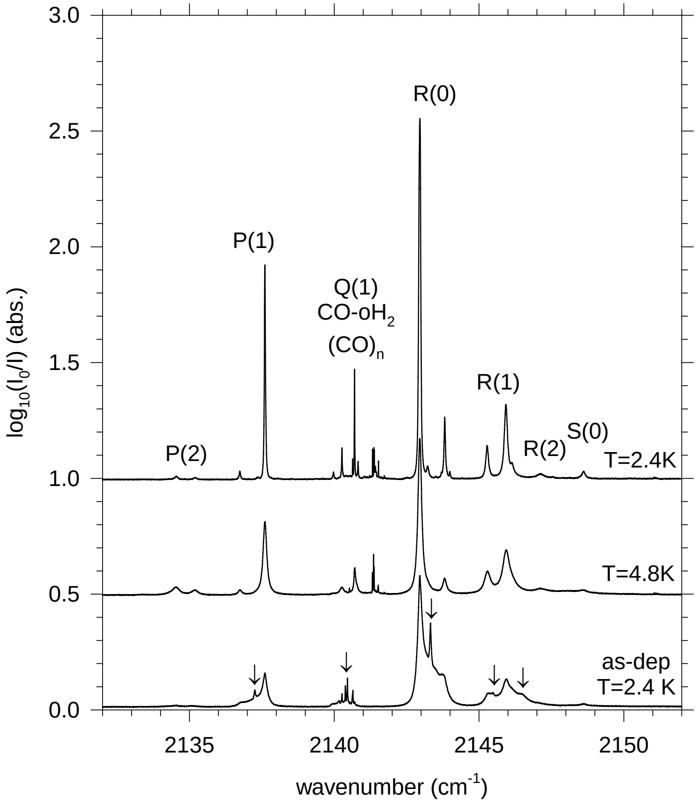
fcc/hcp Regions in RVD Solid pH_2

CH_4/pH_2



S. Tam, M.E. Fajardo, H. Katsuki, H. Hoshina, T. Wakabayashi, and T. Momose, J. Chem. Phys. **111**, 4191 (1999).

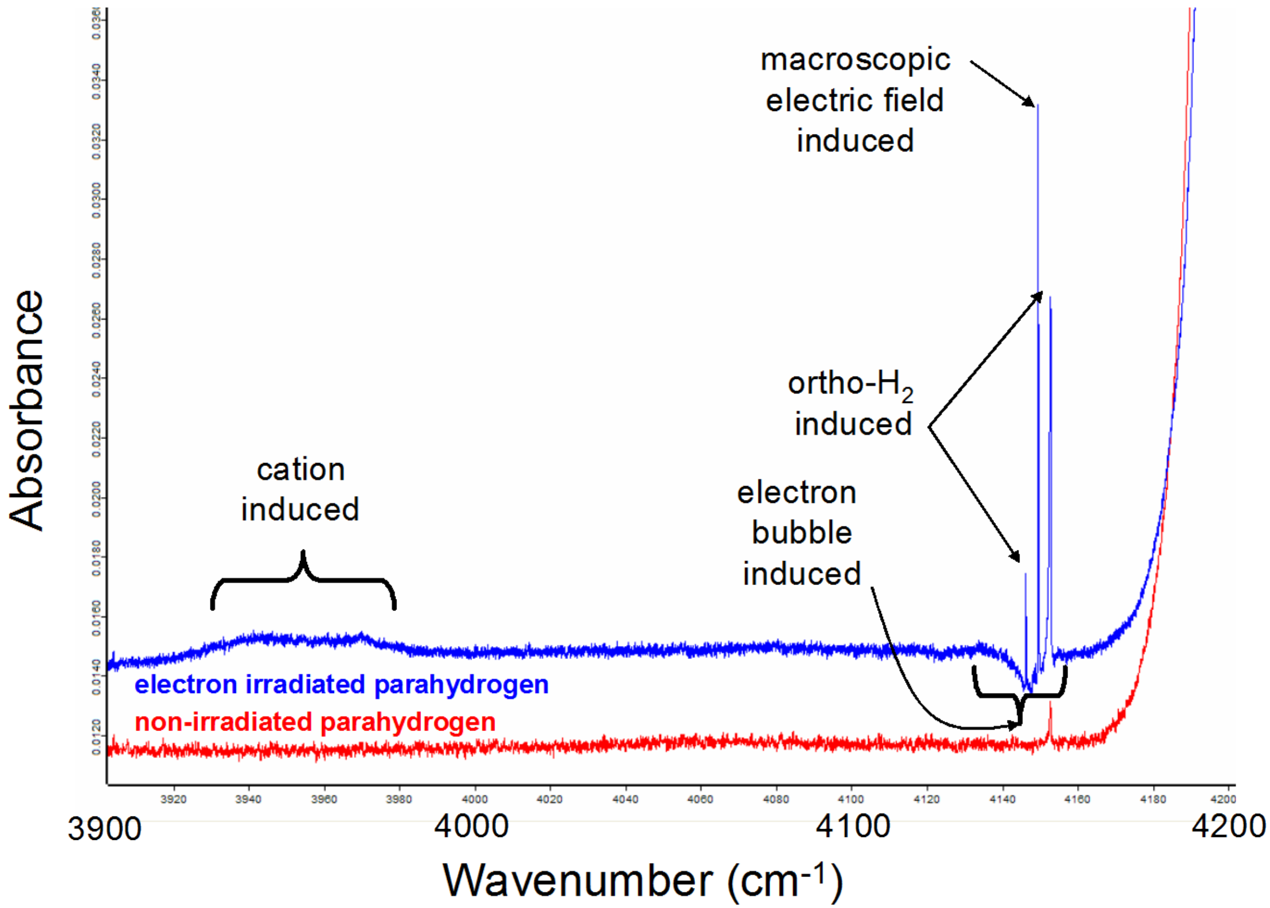
CO/pH_2



M.E. Fajardo, C.M. Lindsay, and T. Momose, J. Chem. Phys. **130**, 244508 (2009).

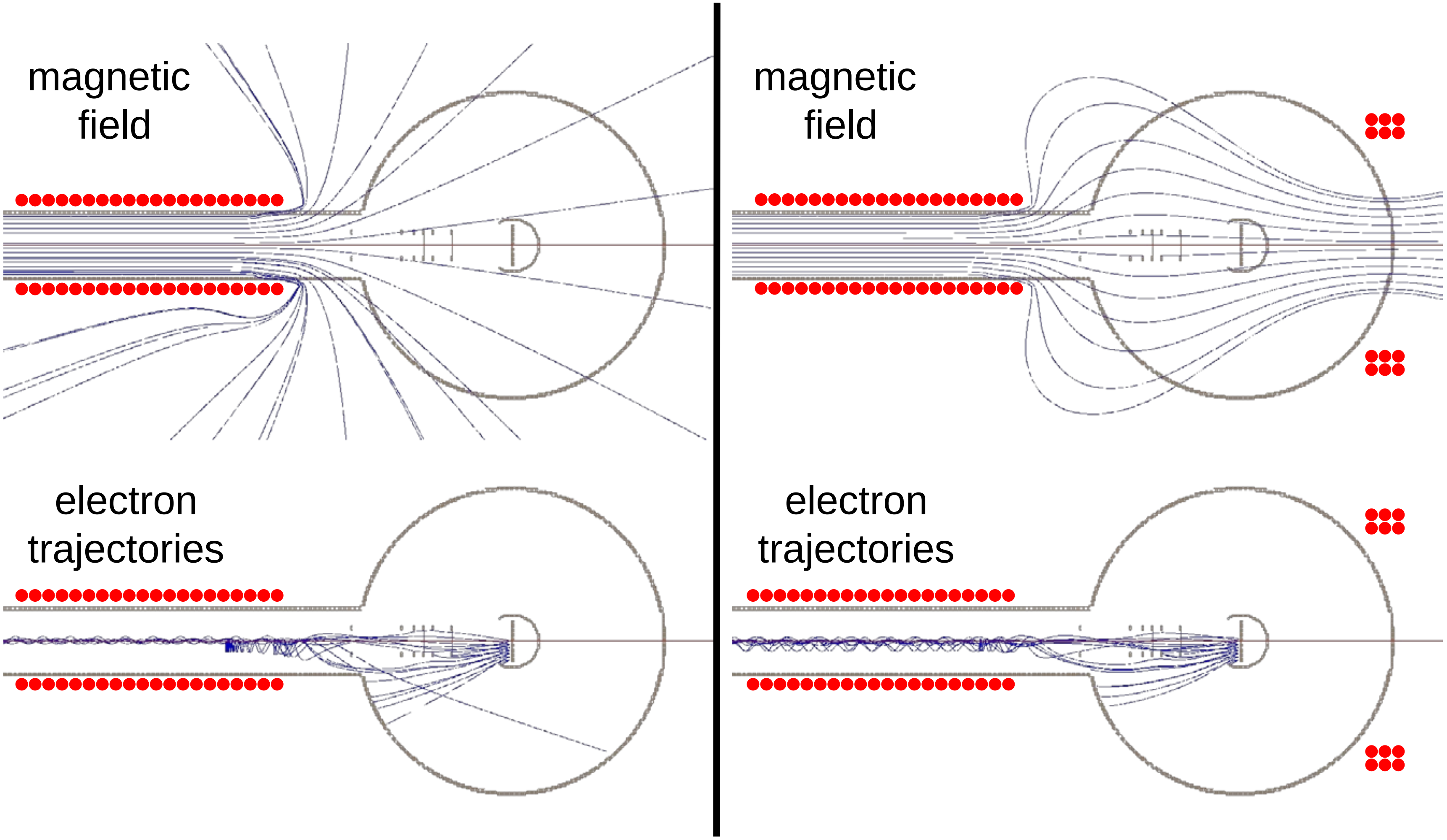
Mixed fcc/hcp microstructure in as-deposited samples; anneals to hcp.

Radiation-Induced Features



Monitor buildup of radiation damage *in-situ* during moderator operation.

Modify Solenoid "Magnetic Funnel"



More electrons collected, but some trajectories evade energy selection gate!

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