



# Modeling Karabut's collimated x-rays, and excess heat in the Piantelli NiH exp't

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Peter Hagelstein

Research Laboratory of Electronics  
MIT

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# CMNS model

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$$\hat{H} = \hbar\omega_0\hat{a}^\dagger\hat{a} + \sum_j \left( \mathbf{M}_j c^2 + \mathbf{a}_j \cdot c\mathbf{P}_j \right)_{\text{stable}} + \sum_j \left( \mathbf{M}_j c^2 + \mathbf{a}_j \cdot c\mathbf{P}_j \right)_{\text{unstable}} - i \frac{\hbar\hat{\Gamma}(E)}{2}$$

Model describes coherent dynamics of stable nuclear states in the presence of a highly excited phonon mode.



# Focus on oscillator coupling with unstable transitions

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$$\hat{H} = \hbar\omega_0\hat{a}^\dagger\hat{a} + \sum_j \left( \mathbf{M}_j c^2 + \mathbf{a}_j \cdot c\mathbf{P}_j \right) - i \frac{\hbar\hat{\Gamma}(E)}{2}$$

unstable

- many ground state nuclei at many sites in the lattice
- relativistic coupling to highly excited states [O(100 MeV)]
- excited states very unstable
- highly-excited phonon mode
- oscillator loss for off-resonance conditions



# Approximate product solution

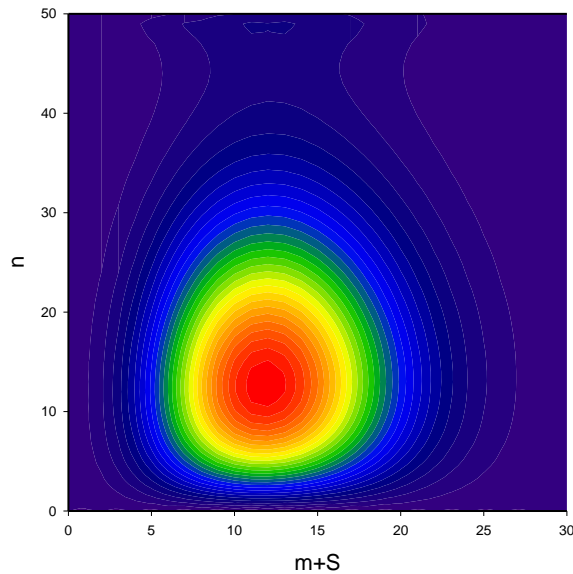
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$$\Psi = \boxed{u_n} \prod_{\beta, \kappa} \left[ (-1)^{N_{\kappa}^{(\beta)}} a_{N_{\kappa}^{(\beta)}}^{(\beta)} \right]$$

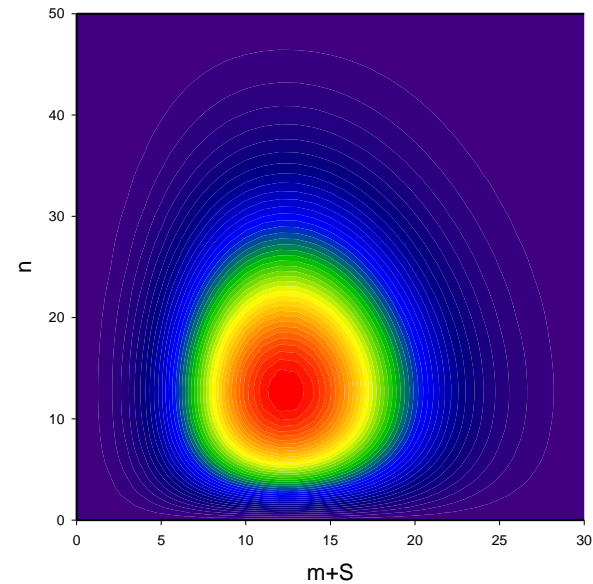
oscillator	excited state
distribution	distributions

Coupling of degrees of freedom is complicated, but we can get a good approximation using a self-consistent Hartree type of approximation

# Two-level system model test problem



exact solution



approximate  
product solution



# Incremental oscillator distribution

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$$u_n \rightarrow u(n) = \text{Ai}\left(\frac{n}{(2g_u)^{1/3}} - 2.83310\right)$$

$$g_u = \sum_{\beta, \kappa} N_1^\beta \frac{2[V_{1\kappa}^{(\beta)}]^2 n_0}{\hbar \omega_0 (E_\kappa^{(\beta)} - E_1^{(\beta)})}$$

Incremental oscillator distribution now available for general case

# Coherent dynamics with two-level systems

oscillator                      two-level systems

$$\begin{aligned}
 \hat{H} = & \boxed{\hbar \omega_0 \hat{b}^\dagger \hat{b}} + \boxed{\Delta E \frac{\hat{S}_z}{\hbar} + V \frac{2\hat{S}_x}{\hbar} (\hat{b} + \hat{b}^\dagger)} \\
 & + \boxed{\sum_j \left( \mathbf{M}_j c^2 + \mathbf{a}_j \cdot c \frac{\partial \mathbf{P}_j}{\partial \mathbf{b}} (\hat{b} + \hat{b}^\dagger) \right)}_{\text{unstable}} \\
 & \boxed{-i \frac{\hbar \hat{\Gamma}(E)}{2}}
 \end{aligned}$$

oscillator loss

N-level systems with  
unstable upper  
states



# Coherent energy exchange

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two-level systems

$$\Psi(t) = \sum_m c_m(t) \left[ |S, m\rangle \Phi_{n_0 - m\Delta n} \right]$$

coupled oscillator and  
unstable transitions

Assuming resonance:

$$\Delta E = \hbar \omega_0 \Delta n$$





# Evolution equation

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$$i\hbar \frac{d}{dt} c_m = \langle \Phi_{n_0} | b + b^\dagger | \Phi_{n_0 + \Delta n} \rangle \sqrt{(S + m)(S - m - 1)} c_{m-1} \\ + \langle \Phi_{n_0} | b + b^\dagger | \Phi_{n_0 - \Delta n} \rangle \sqrt{(S - m)(S + m - 1)} c_{m+1}$$

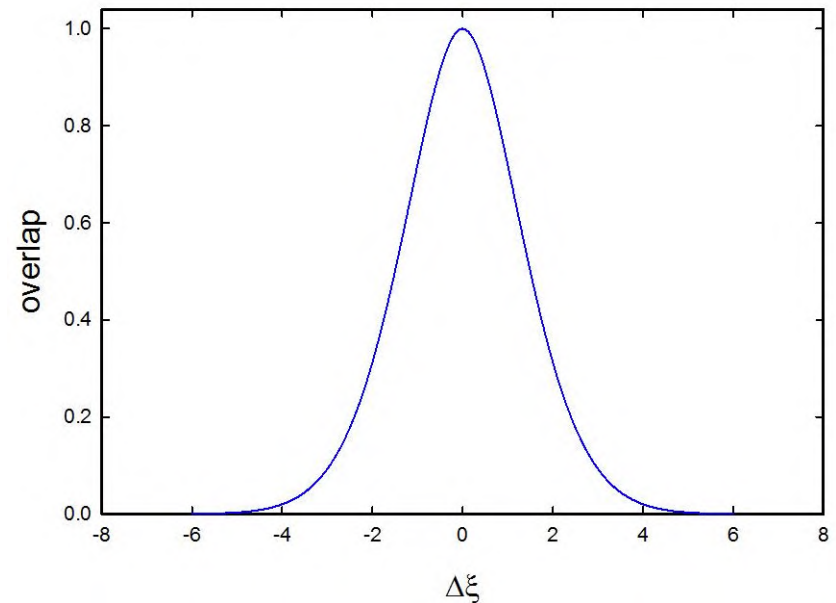
Model describes coherent energy exchange between two-level systems and oscillator (coupled to N-level systems).

Whether significant energy exchange occurs depends on phonon matrix elements

# Phonon matrix element

$$\frac{1}{2} \langle \Phi_{n_0} | \hat{b} + \hat{b}^\dagger | \Phi_{n_0 + \Delta n} \rangle = \frac{\int_{-2.83310}^{\infty} \text{Ai}(\xi) \text{Ai}(\xi + \Delta\xi) d\xi}{\int_{-2.83310}^{\infty} \text{Ai}^2(\xi) d\xi} = f(\Delta\xi)$$

$$\Delta\xi = \frac{\Delta n}{(2g_u)^{1/3}}$$





# Assessment

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- Coherent energy exchange between (stable) two-level systems and oscillator (coupled to N-level systems)
- Coupled oscillator and N-level systems fractionates large quantum
- Stronger coupling  $\leftrightarrow$  larger  $g_u \leftrightarrow$  more quanta can be exchanged
- No exchange in absence of oscillator loss effects



# Can we go the other way?

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- Donor receiver model developed for converting donor energy to oscillator excitation
- But can we go the other way?
- Is it possible to start with an excited oscillator, and transfer the energy to two-level systems
- Is it possible to excite nuclei with vibrational excitation?
- New models would say yes



# Can it be demonstrated?

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- Model predicts coherent energy exchange between equivalent two-level systems with a large transition energy, and an oscillator with a low transition energy
- Two-level systems can be atoms, molecules, nuclei, spin systems
- Oscillator can be vibrational, electrical, plasmonic, electromagnetic
- Many possibilities for systems to show the effect
- But can be demonstrate it between phonons and nuclei?



# Critical parameters

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$$\Delta\xi = \frac{\Delta n}{(2g_u)^{1/3}} \quad \text{needs to be near unity}$$

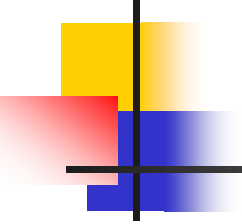
$$g_u = \sum_{\beta, \kappa} N_1^\beta \frac{2[V_{1\kappa}^{(\beta)}]^2 n_0}{\hbar \omega_0 (E_\kappa^{(\beta)} - E_1^{(\beta)})}$$

Want  $\Delta n$  to be small; want  $\omega_0$  to be large, want low energy transition, need highly excited oscillator

# What are lowest energy nuclear transitions?

Nucleus	Excited state energy (keV)	half-life	multipolarity
$^{201}\text{Hg}$	1.5648	81 ns	M1+E2
$^{181}\text{Ta}$	6.240	$6.05 \mu\text{s}$	E1
$^{169}\text{Tm}$	8.41017	4.09 ns	M1+E2
$^{83}\text{Kr}$	9.4051	154.4 ns	M1+E2
$^{187}\text{Os}$	9.75	2.38 ns	M1(+E2)
$^{73}\text{Ge}$	13.2845	$2.92 \mu\text{s}$	E2
$^{57}\text{Fe}$	14.4129	98.3 ns	M1+E2

P. L. Hagelstein, "Bird's eye view of phonon models for excess heat in the Fleischmann-Pons experiment," *J. Cond. Mat. Nucl. Sci.* (in press)



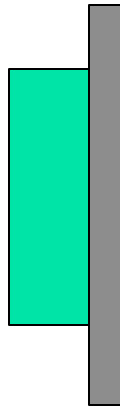
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$^{201}\text{Hg}$  transition at 1565 eV is optimum candidate among stable nuclei to demonstrate effect.



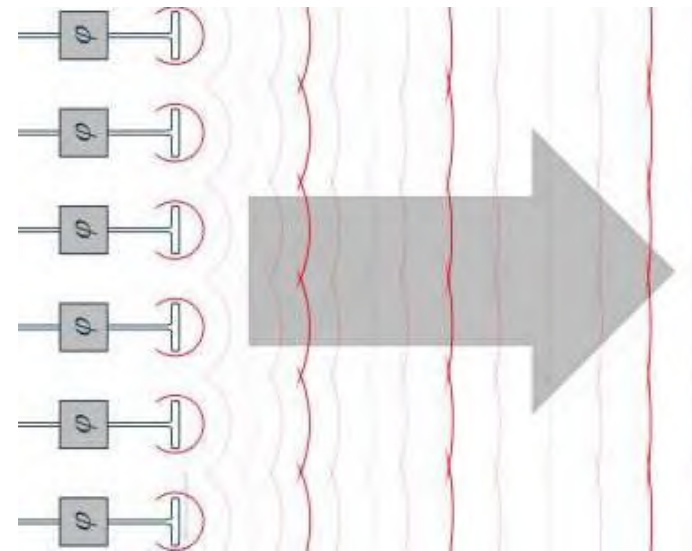
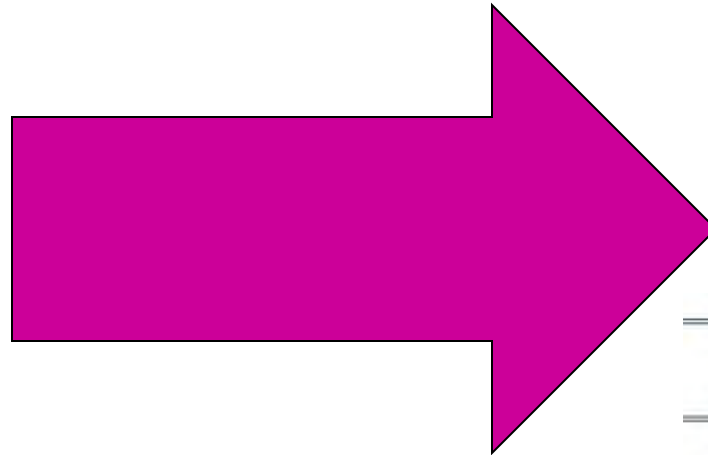
# Conceptual design

ACME THz  
vibrational  
source

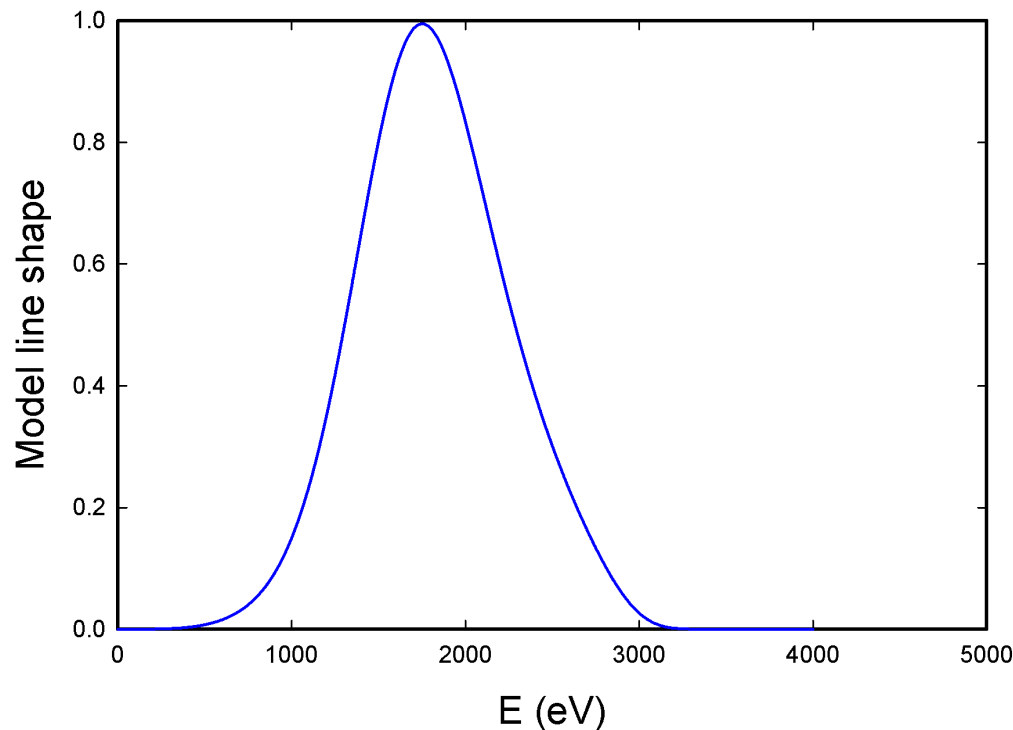


Hg containing  
sample

1.5 keV collimated x-rays



# Predicted spectrum is broad



Spread in phonon distribution that causes excitation will show up in the broadening of the line, and shift due to  $E^2$  in photon density of states.

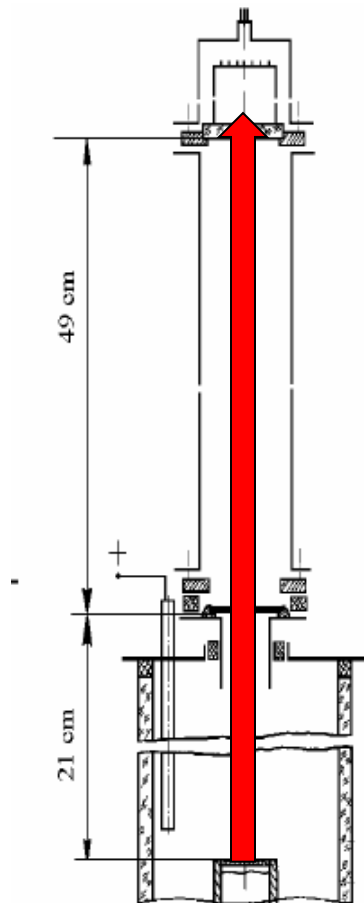


# Karabut experiment

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- Alexander Karabut (Luch Institute, Moscow) showed up at ICCF10 talking about having demonstrated an x-ray laser
- Karabut was working with a high-current glow discharge
- Karabut observed collimated x-ray radiation near 1.5 keV
- (No way for there to be an x-ray laser in this experiment)
- Must be some alternate explanation...

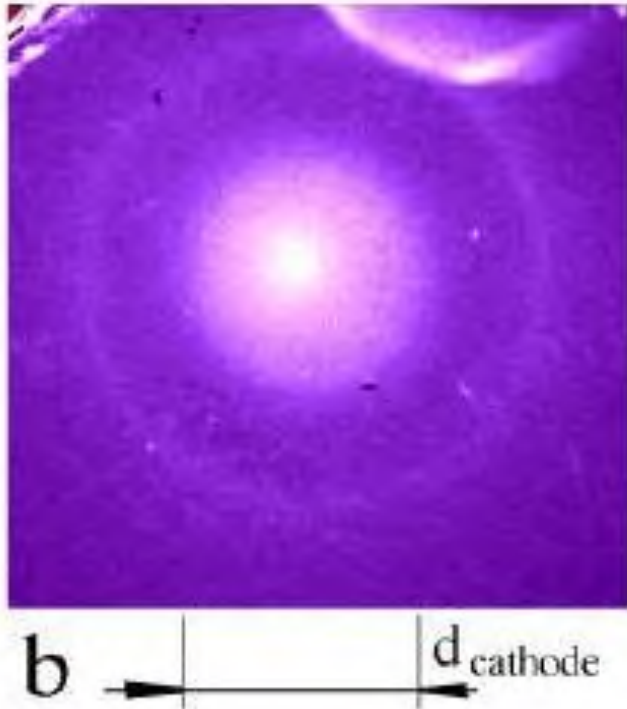
# Karabut experiment (ICCF10)



Collimated x-ray effect seen  
with different metals (Al, V, Fe,  
Zn, Mo, Pd, W, others)

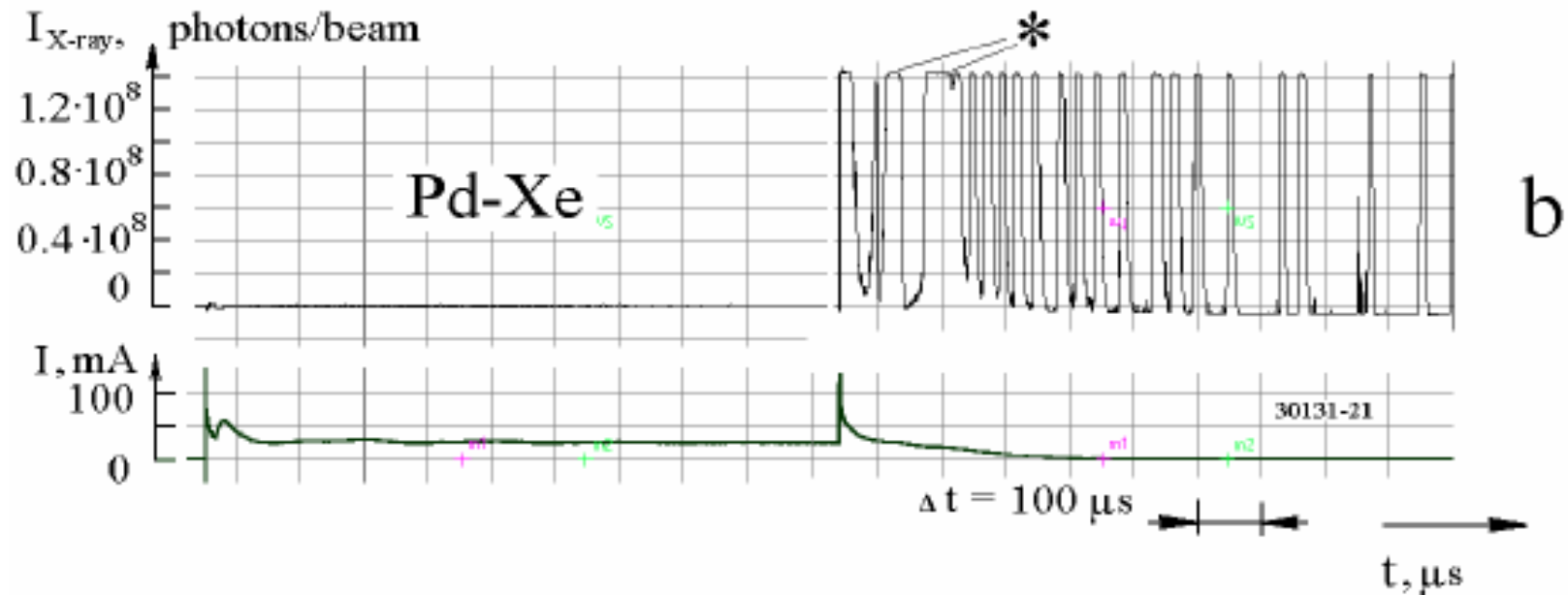
...and with different gasses ( $H_2$ ,  
 $D_2$ , Kr, Xe)

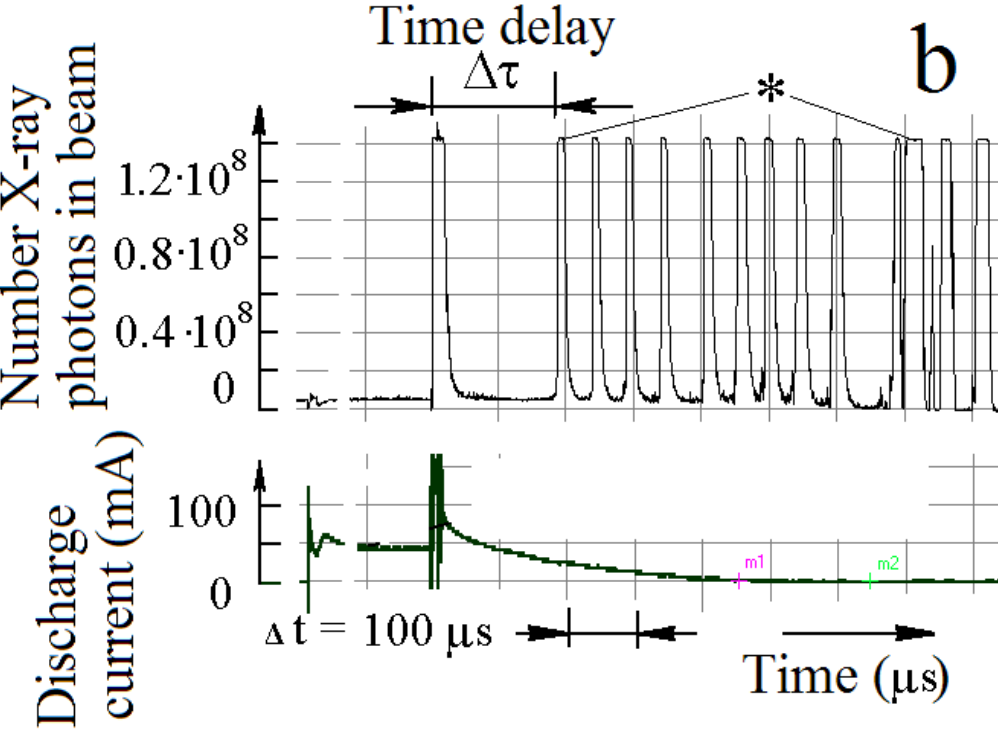
# Pinhole camera image of cathode



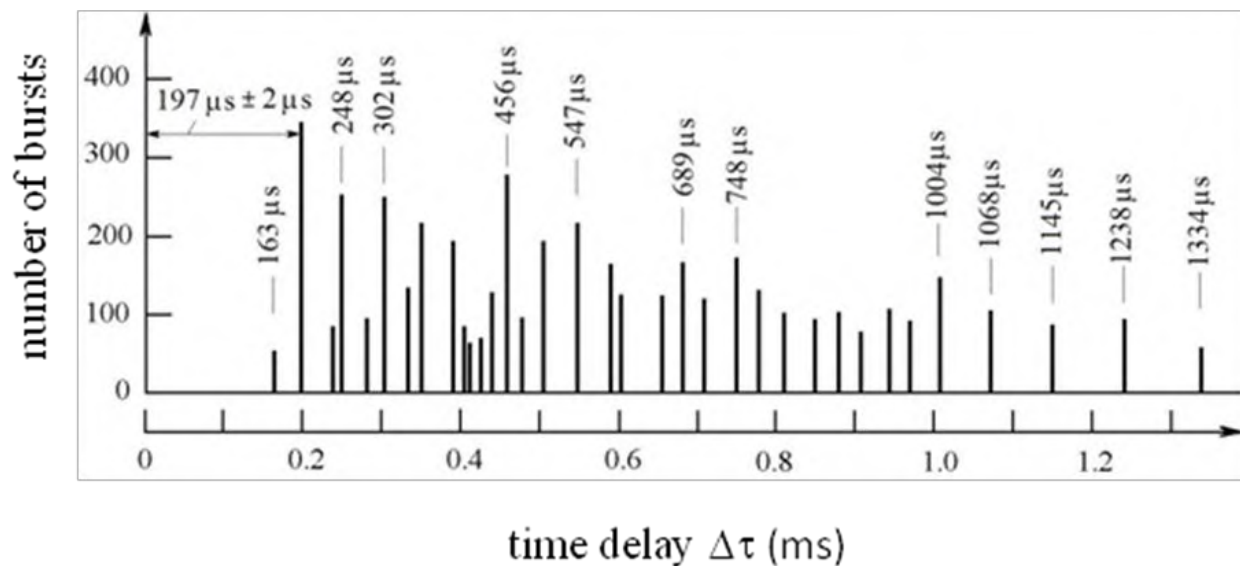
Collimated x-rays very bright,  
originate from cathode surface

# Emission after turn off





A. B. Karabut, E. A. Karabut, P. L. Hagelstein, "Spectral and temporal characteristics of x-ray emission from metal electrodes in a high-current glow discharge," *J. Cond. Mat. Nucl. Sci.* (in press).



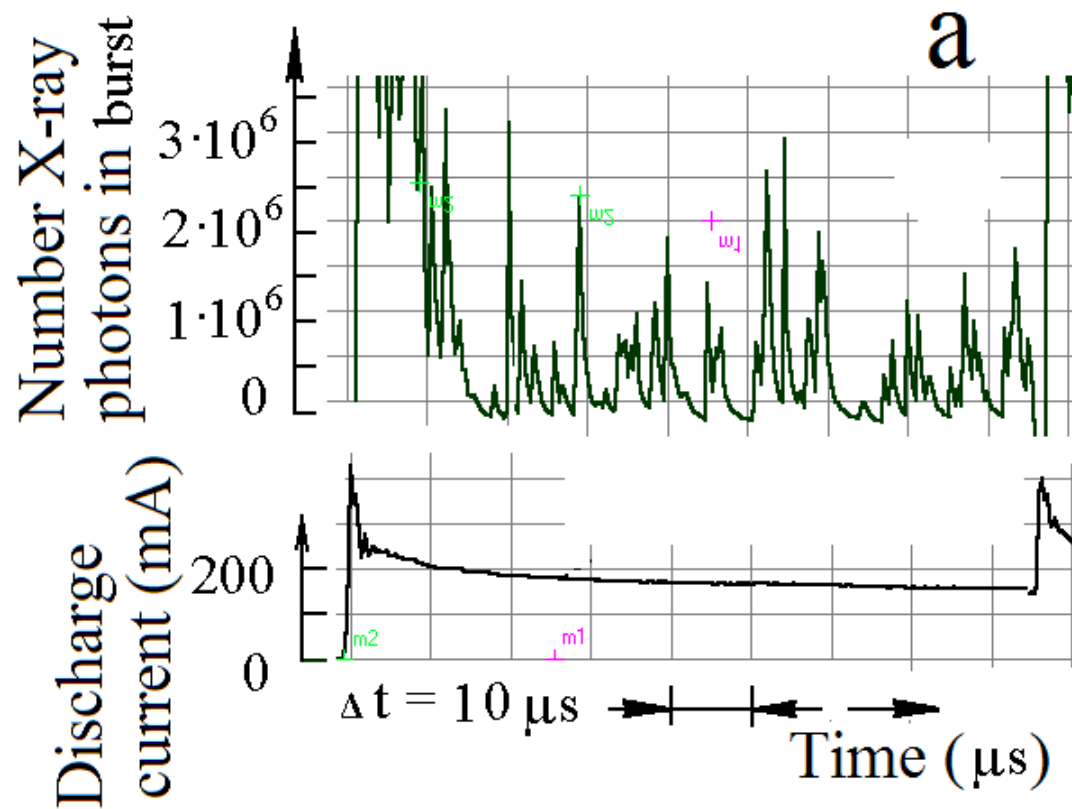


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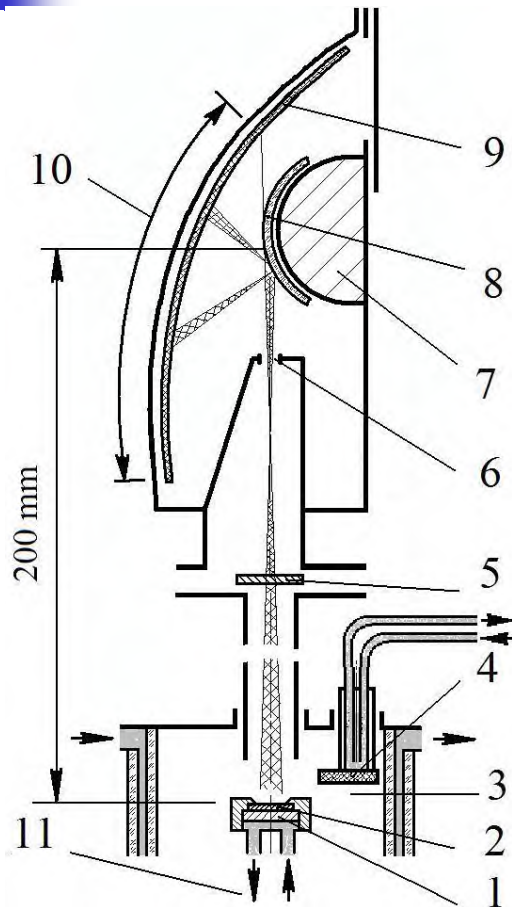
Collimated emission appears after discharge is turned off,  
up to 1 msec and more



# Diffuse emission during discharge



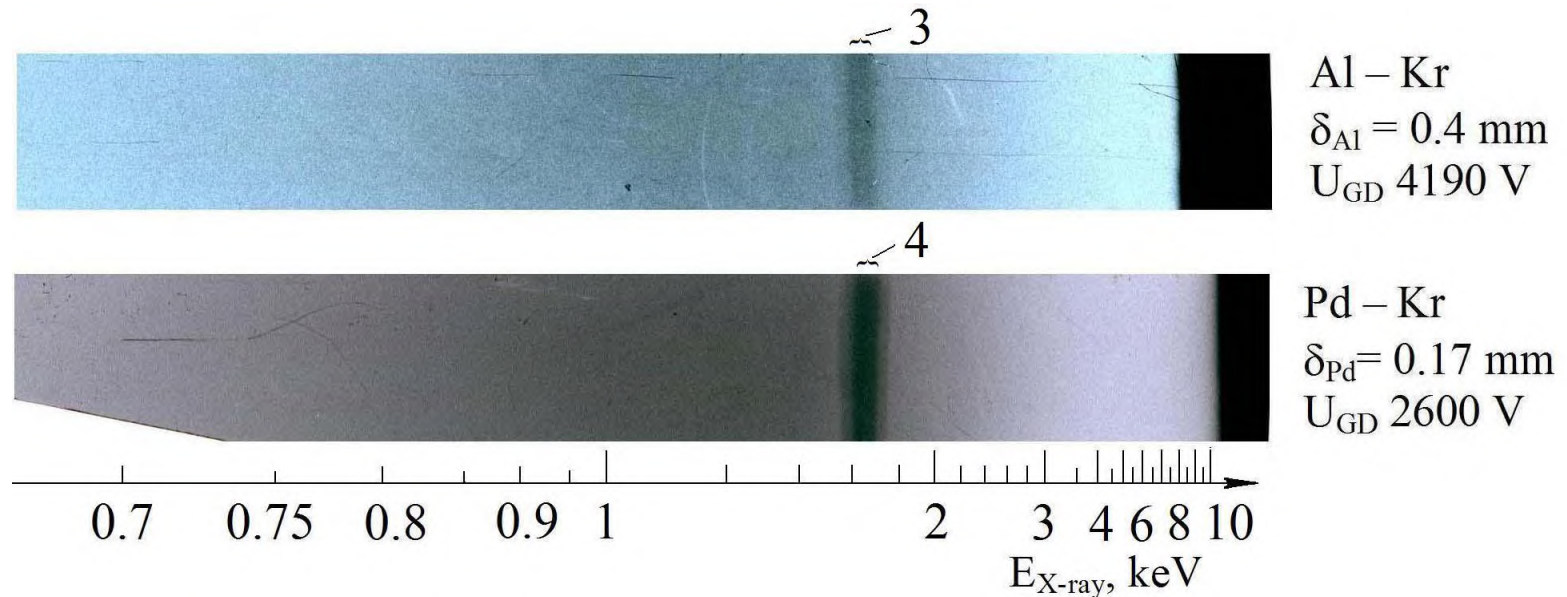
# Bent mica spectrometer



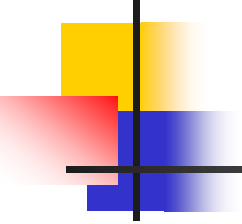
## **X-ray spectrometer:**

- 1– cathode holder,
- 2– cathode sample,
- 3 – vacuum discharge chamber,
- 4 – anode,
- 5 –  $15\ \mu$  Be screen ,
- 6 – input slit of spectrometer,
- 7 – crystals holder,
- 8 – curved mica crystal,
- 9 – x-ray film,
- 10 – area of reflection spectra ,
- 11 – input and output cooling water.

# Diffuse Kr L-shell emission



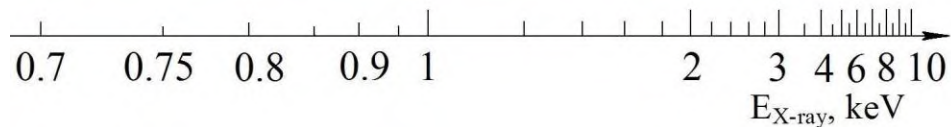
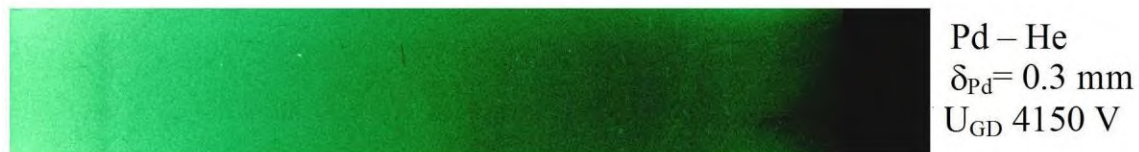
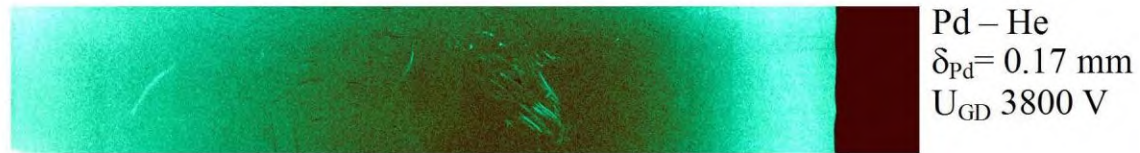
X-ray spectra for Pd and Al cathodes taken in Kr gas showing characteristic L-shell emission (denoted as 3 and 4 in the spectra) near 1.6 keV (the  $L_{\alpha 1}$  and  $L_{\alpha 2}$  transitions are listed at 1.581 keV and at 1.580 keV). Minor differences between the observed and known energy may be due to the use of the normal incidence grating formula.



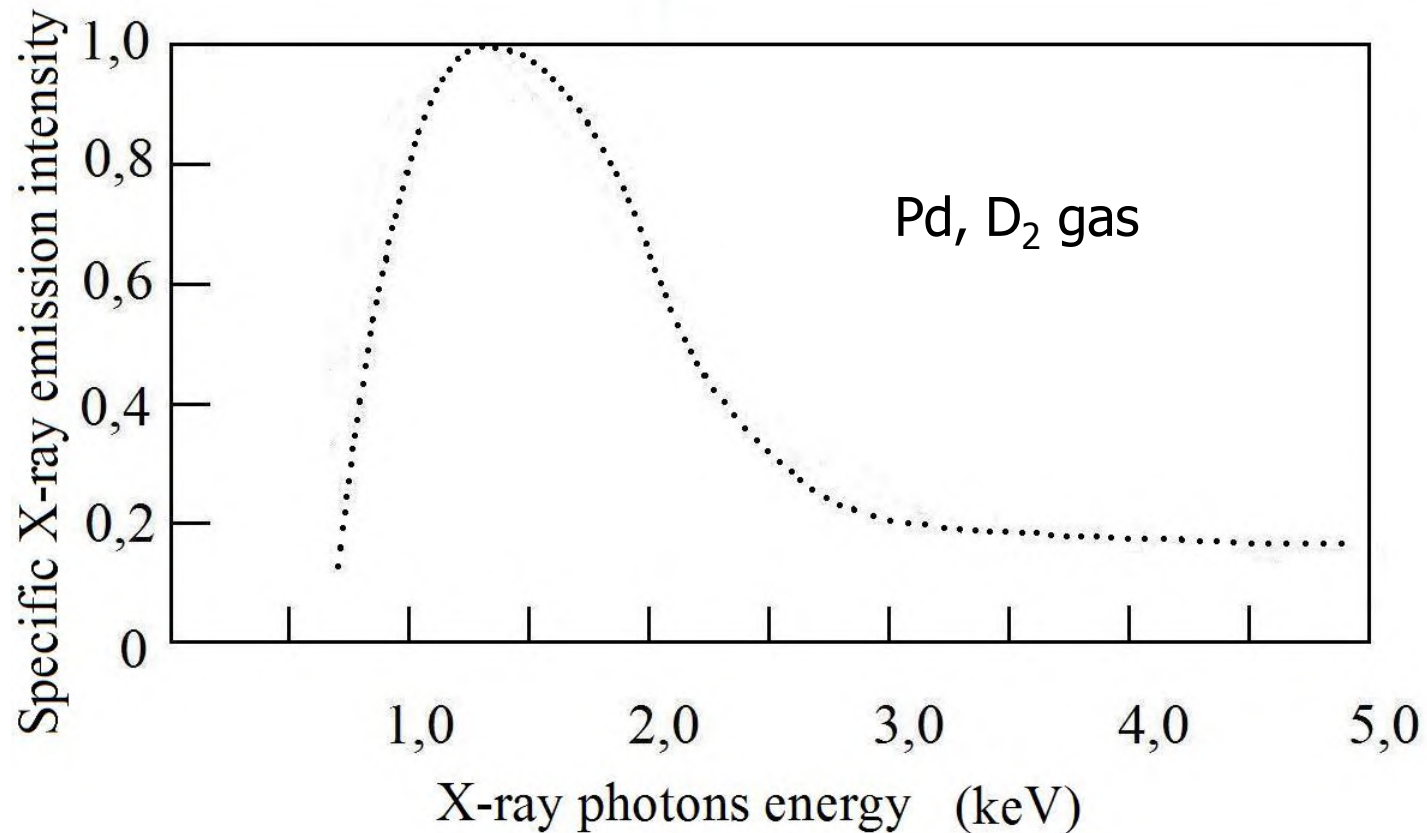
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The Kr L-shell line is diffuse and originates from the cathode surface (not from the gas). It shows up in the spectrum a bit off due to the way the data was analyzed.

# Diffuse continuum emission

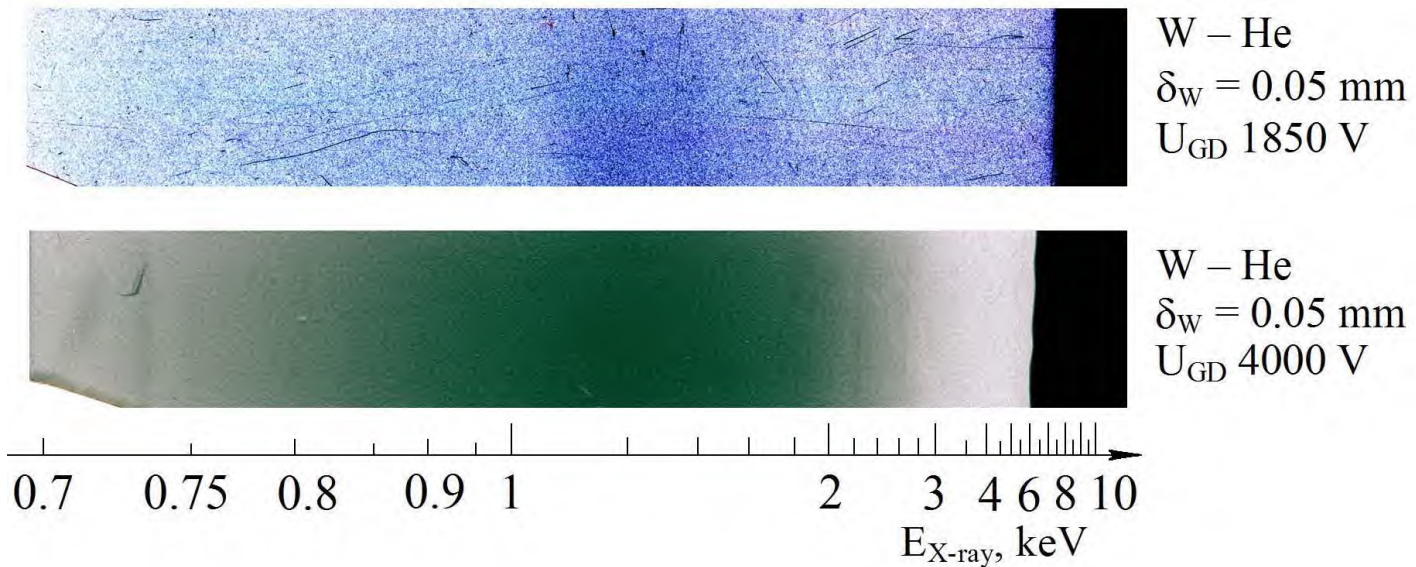


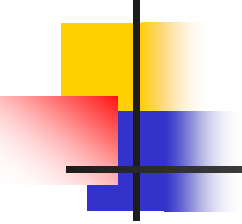
# Spectrum of continuum





# Voltage dependence



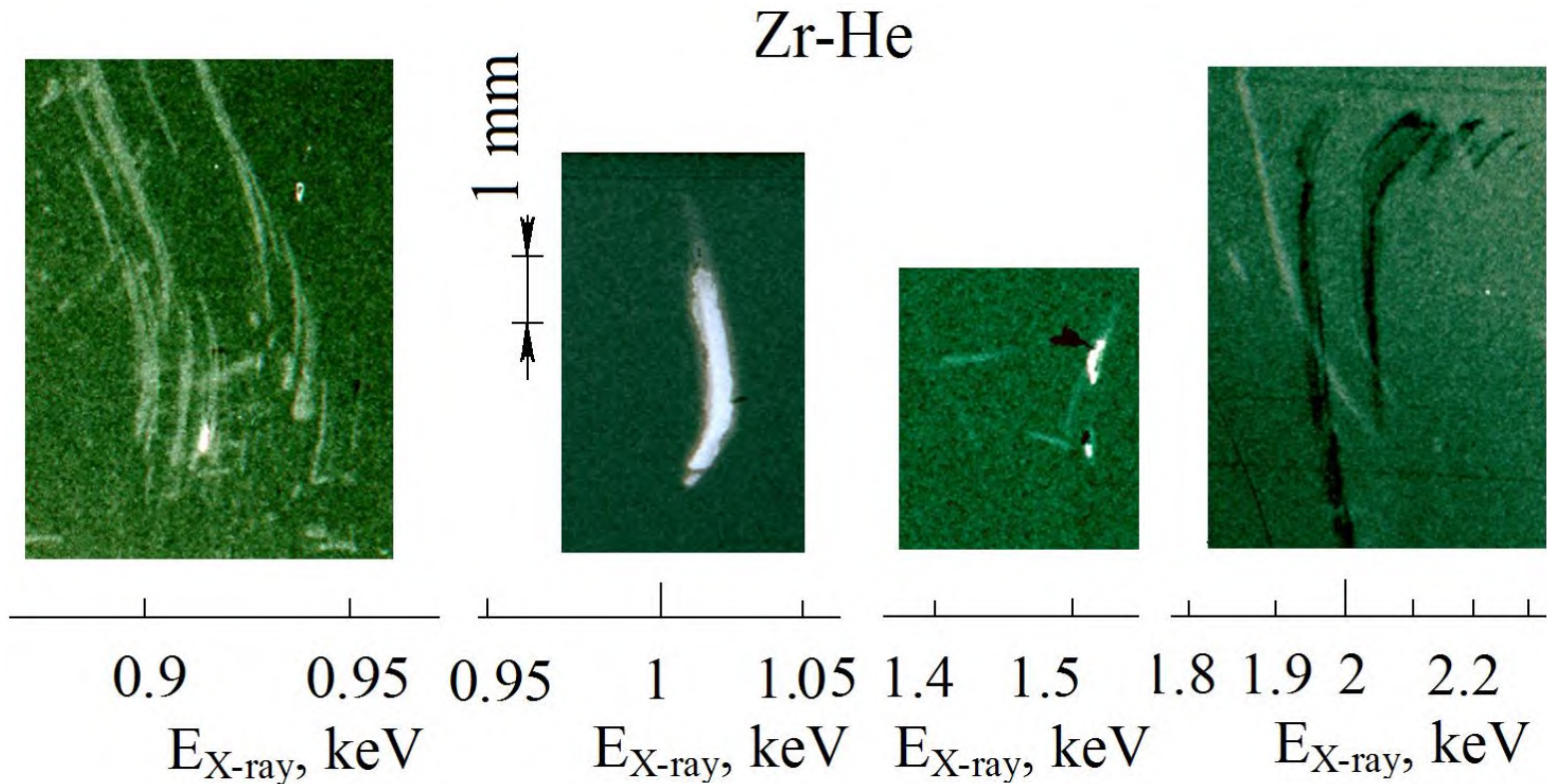


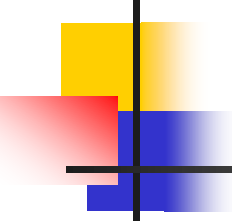
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The diffuse continuum emission is a broad feature that originates from the cathode surface. The width depends on the applied voltage (and hence current).



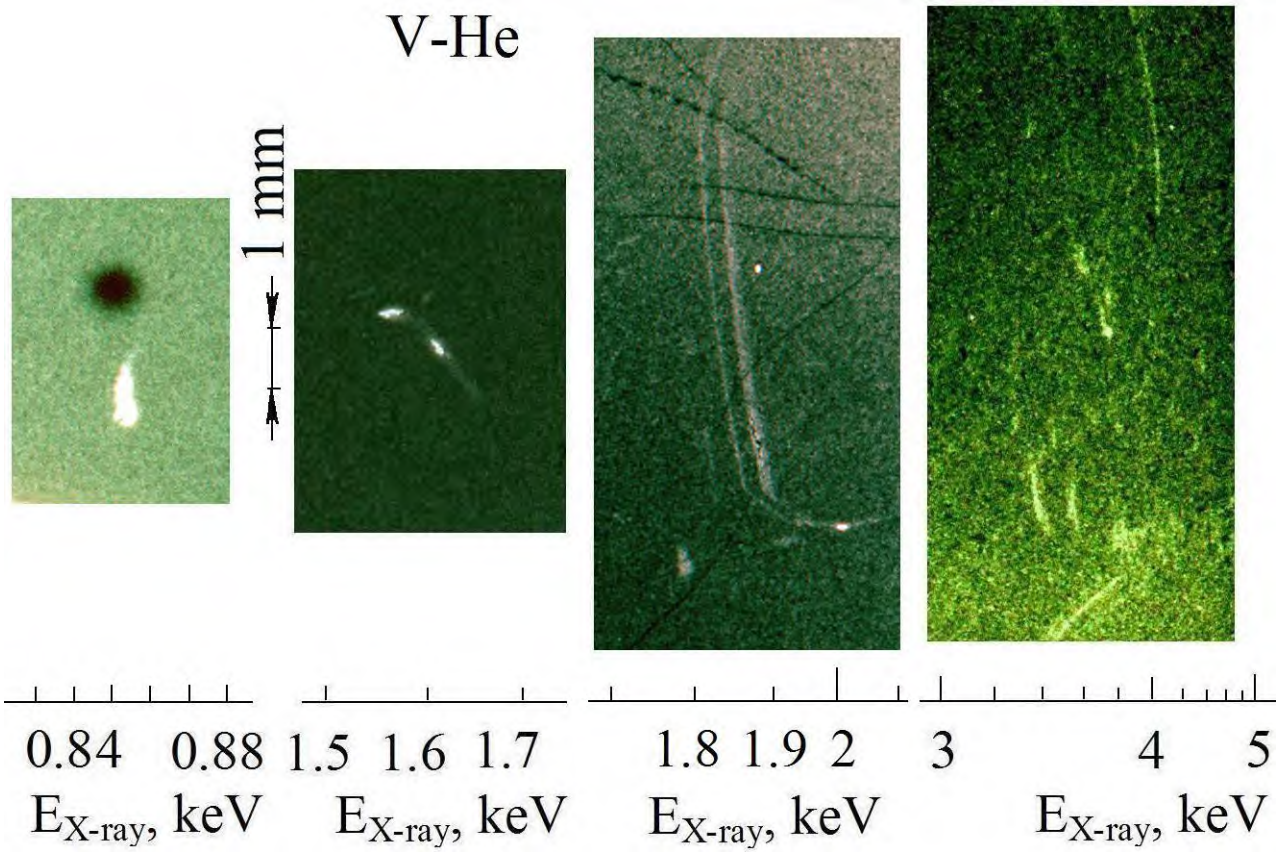
# Collimated beamlets



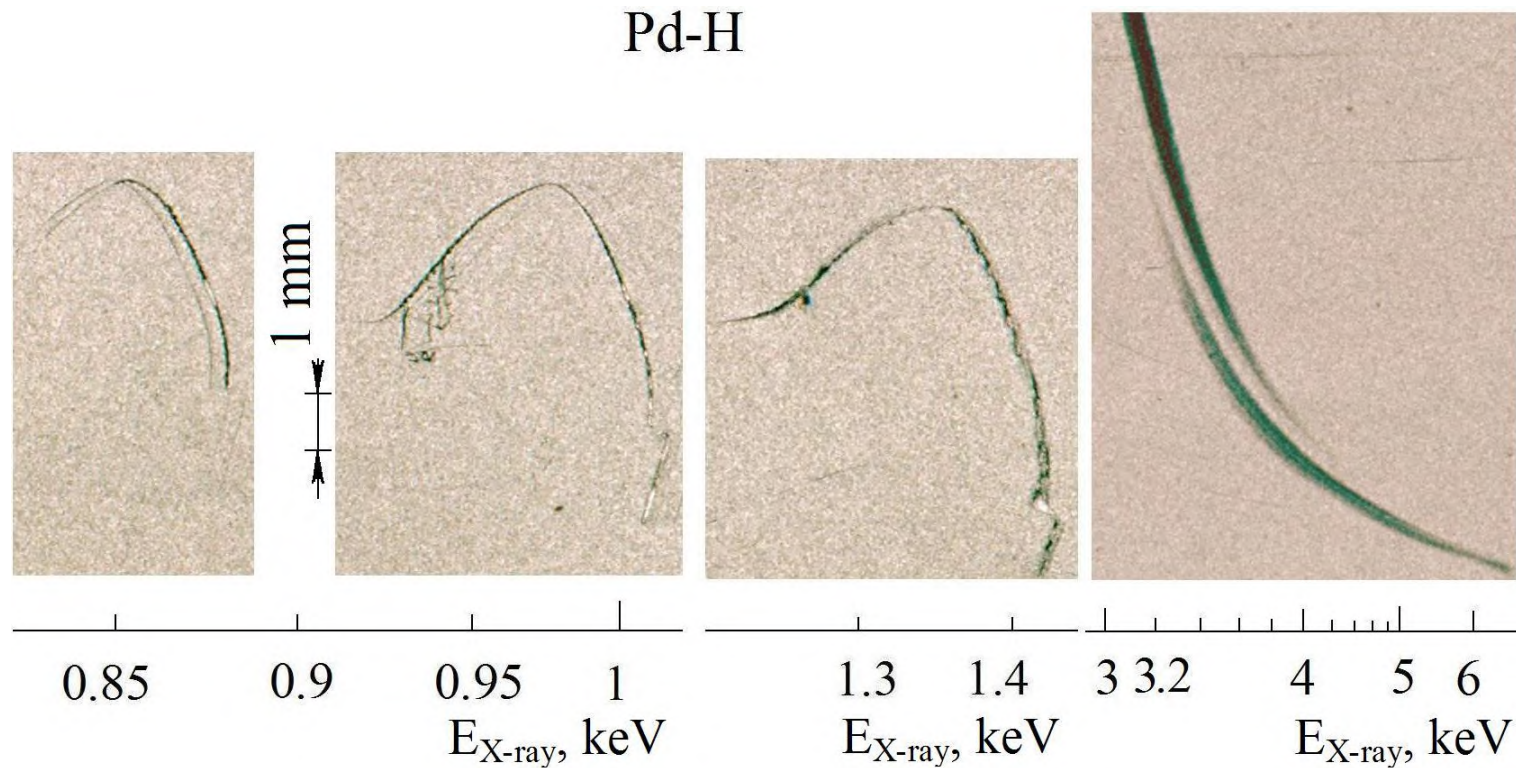


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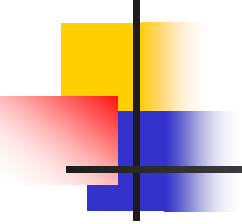
Interpret the curves as due to a minor change in direction of the beam during the emission. The emission is sufficiently bright to damage (cause solarization) film.



# Beams seen long after discharge



Data collected for 20 hours after the discharge turned off



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Collimated emission taken long after the discharge is turned off is particularly interesting because effect can only be due to vibrational effects (no possible residual from the discharge).



# Collimated emission

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- Collimated emission due to coherent energy exchange from O(50 MHz) vibrational modes
- Hg sputtered into surface from gas contamination by discharge
- Phase coherence only possible for narrow (and almost random) frequencies within broad line width due to random positions of Hg nuclei [based on simulations]



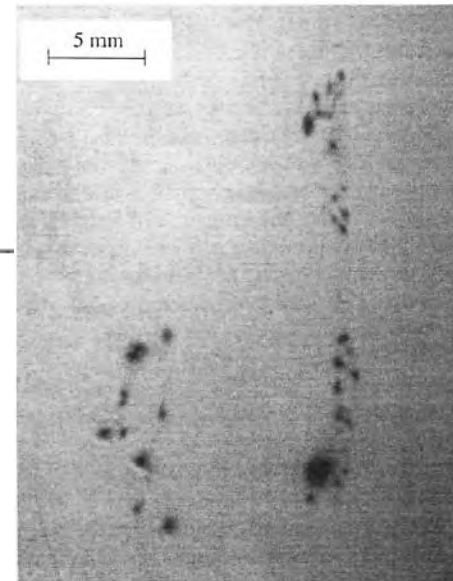
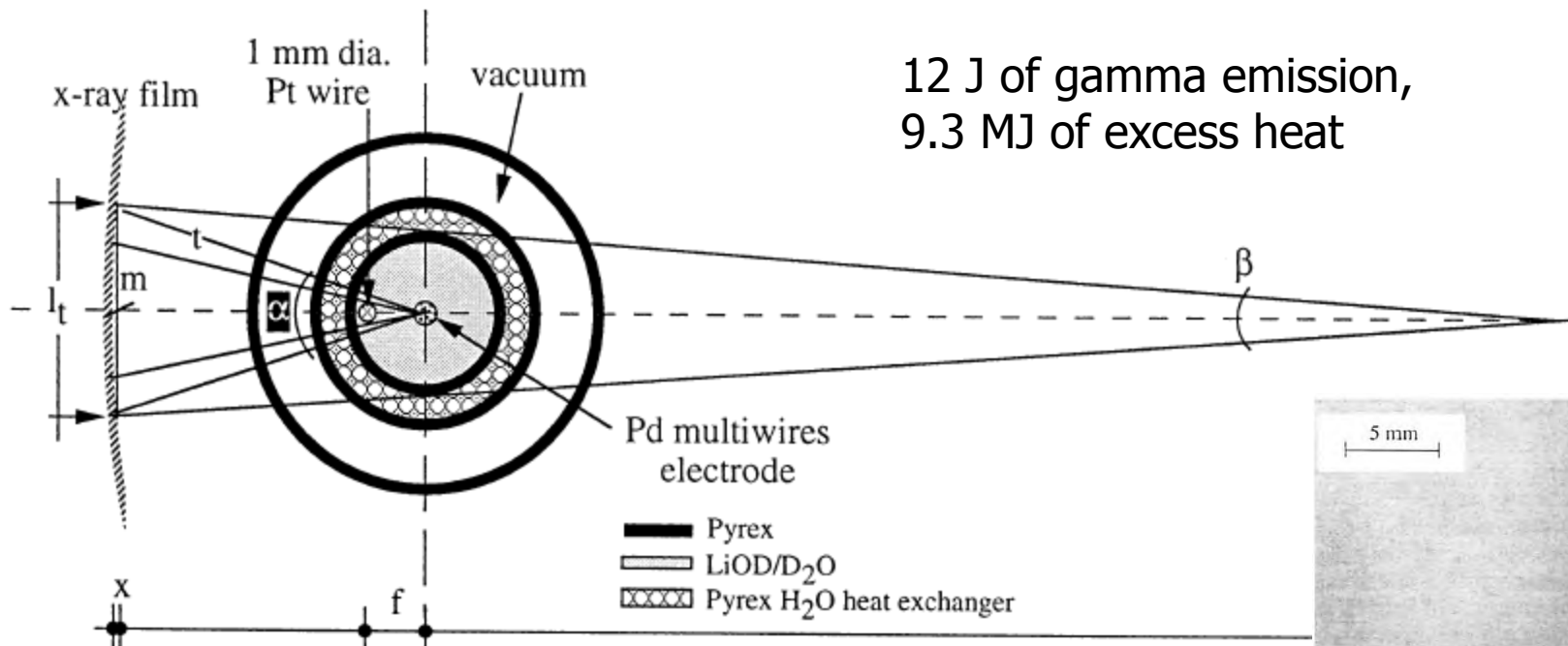


# Diffuse emission

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- Diffuse emission due generally to coherent energy exchange with THz phonon modes excited by discharge ion bombardment
- Broad feature due to  $^{201}\text{Hg}$  nuclear transition
- Characteristic gas line emission due to energy exchange with  $^{201}\text{Hg}$  excited state (Kr, Ar correlated in space with Hg)
- Characteristic host line emission due to similar energy exchange effect

# Gozzi's experiment (ICCF6)



From absorption coefficient of Pd cathodes, can estimate the energy of the gamma signal. Gozzi et al obtained an energy of 89 keV, and suggested that it was due to  $^{109m}\text{Ag}$





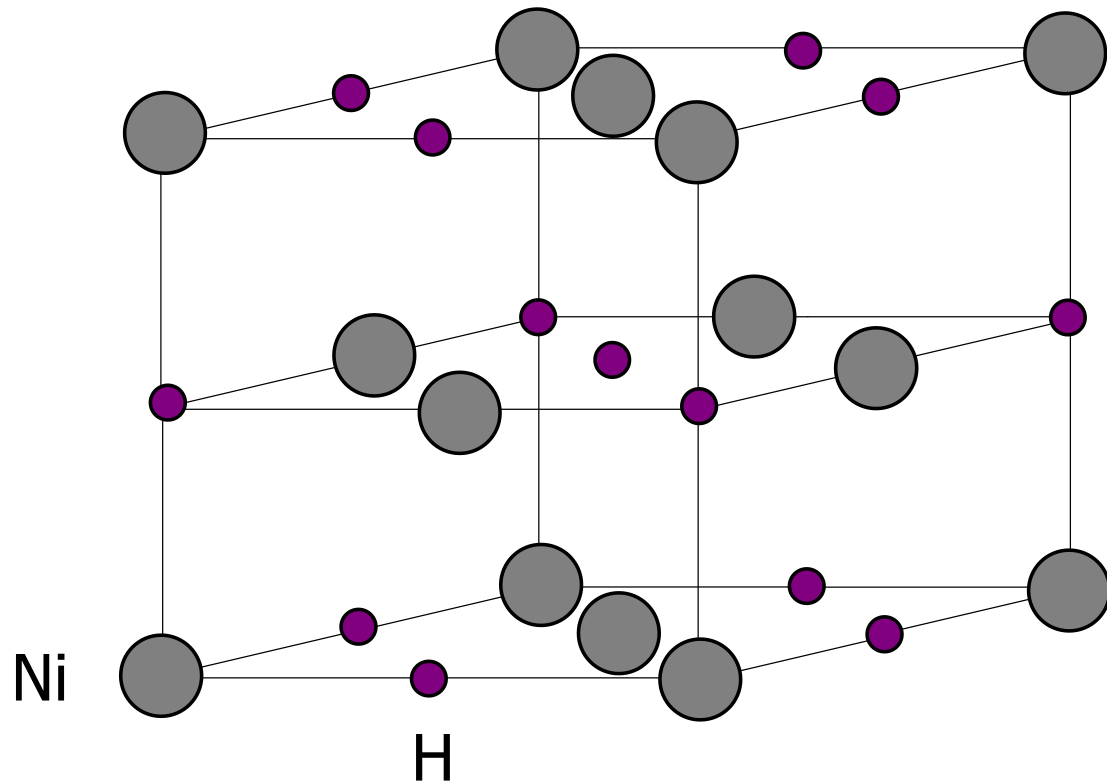


# Before looking at expts...

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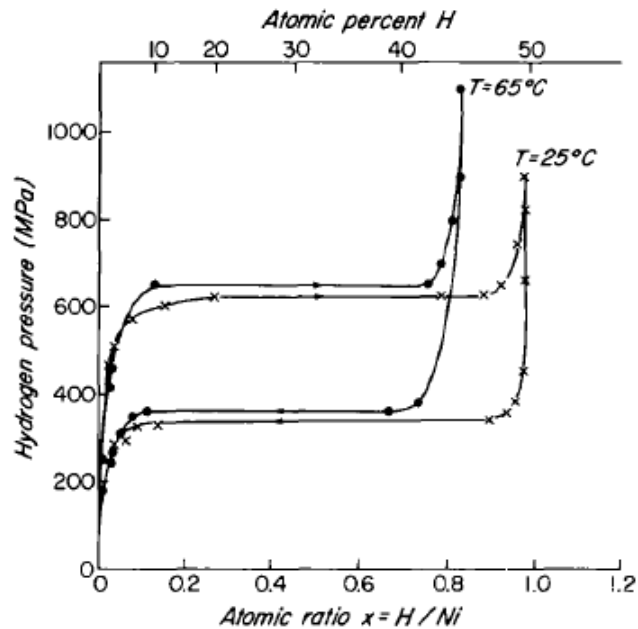
- Excess heat seen in NiH experiments
- Effect first reported in electrolysis experiments by Mills and Kneizys (1991)
- Excess heat in gas loading experiments reported by Piantelli et al (1994)
- NiH is not PdD
- Differences are important

# NiH lattice structure (fcc)



# Phase diagram

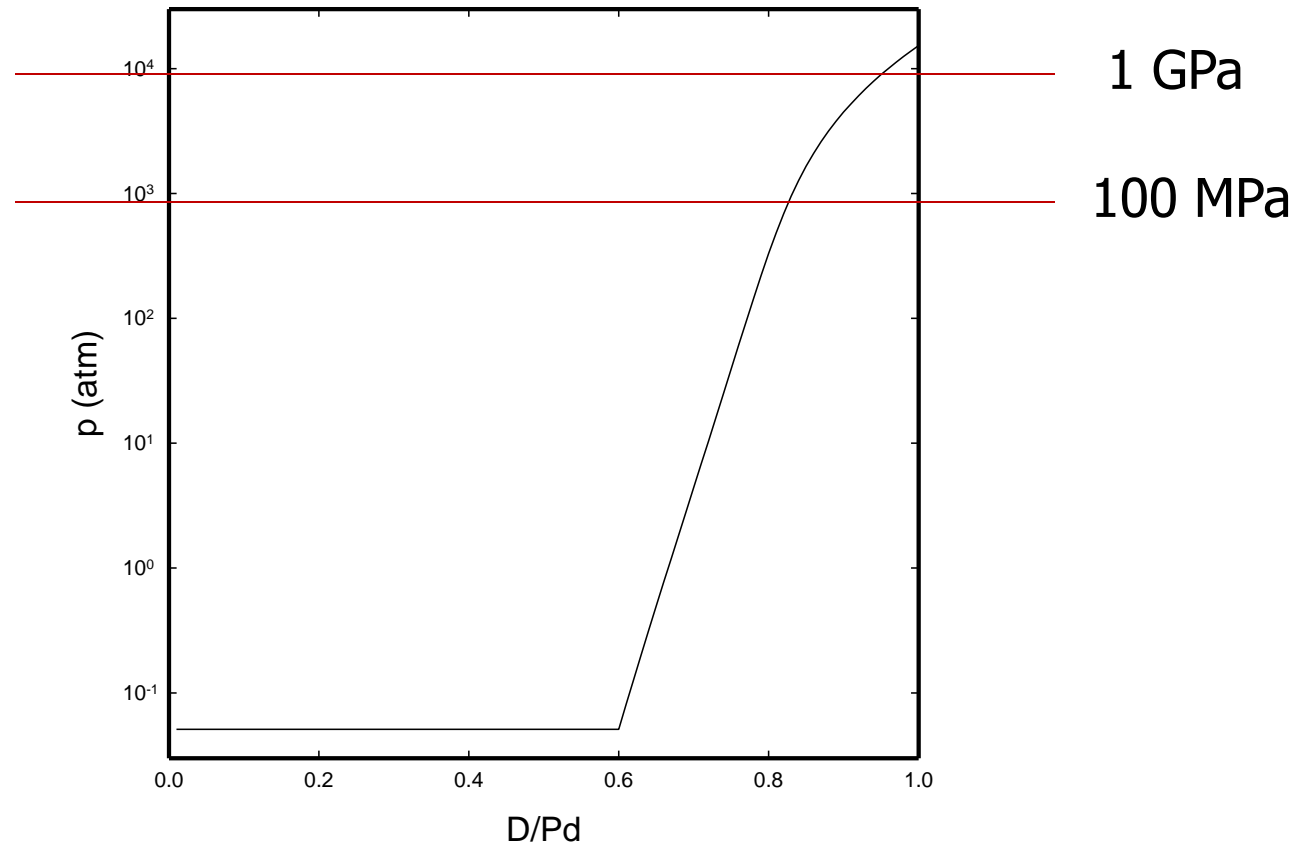
**Fig. 4 Absorption and Desorption Isotherms at 25 and 65 °C for the Ni-H System**



From [78Bar]. M. Wayman and G.C. Weatherly, 1989.

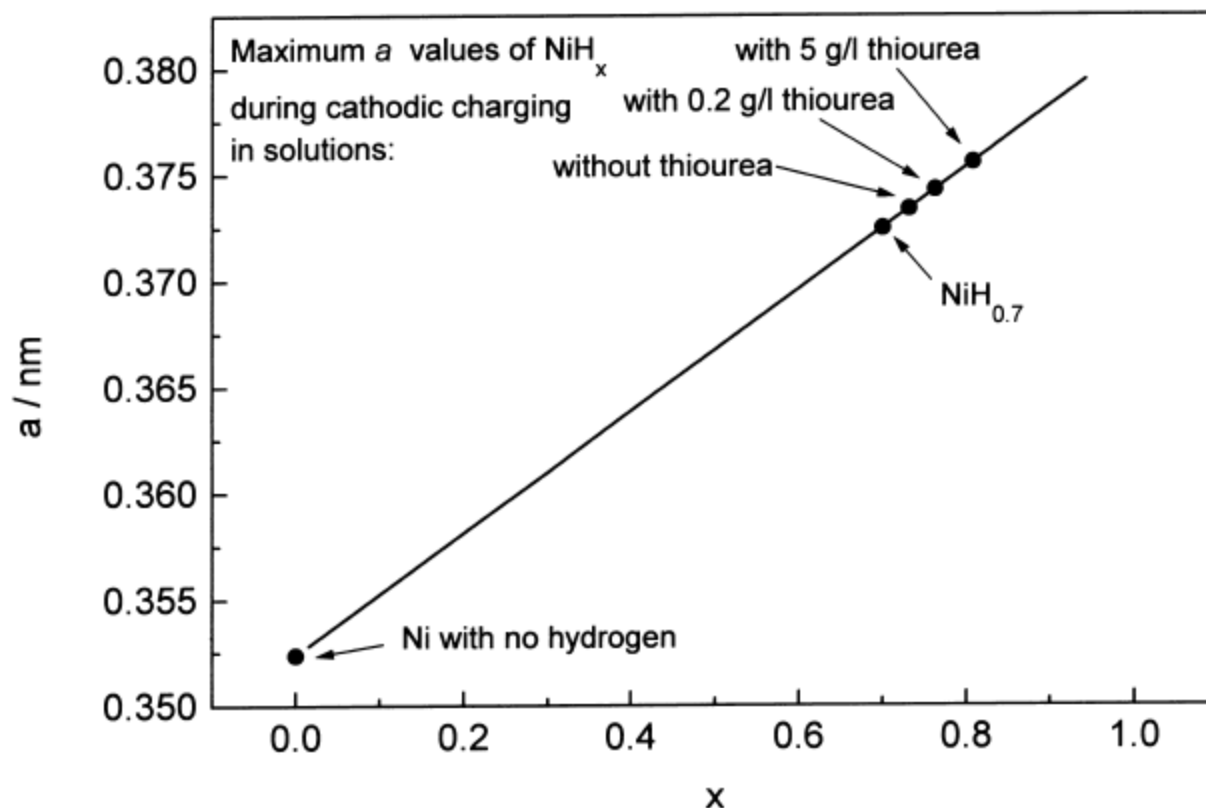
Note: 400 Mpa = 3948 atm

# Pressure vs loading



# X-ray diffraction data in electrochemical loading

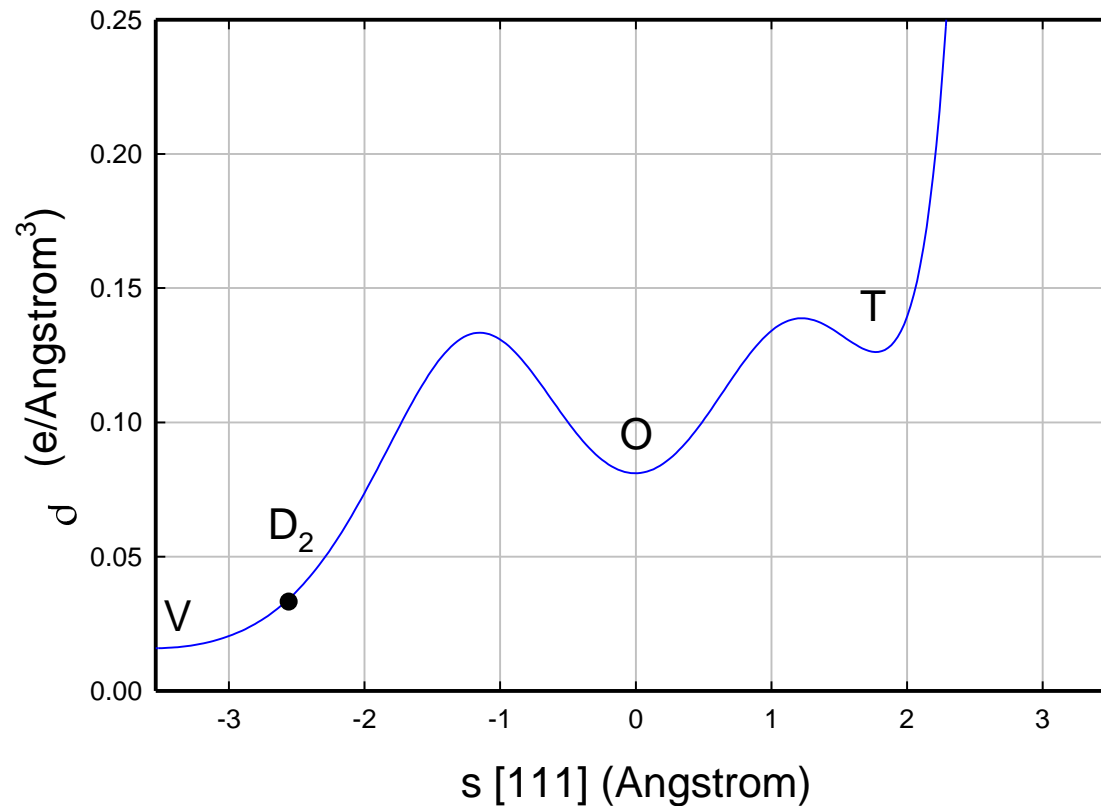
Juskenas et al, *Electrochimica Acta* **43** 1903 (1998)



No evidence for intermediate  $a$  values for loadings in the  $\alpha$ - $\beta$  phase region. Observed only is the change in volume occupied by  $\beta$ -phase  $\text{NiH}$ .

Fig. 8. Dependence of calculated (solid line) and experimental (●) values of the lattice parameter  $a$  of  $\beta$ - $\text{NiH}_x$  phase on the stoichiometric coefficient  $x$  of the hydride phase.

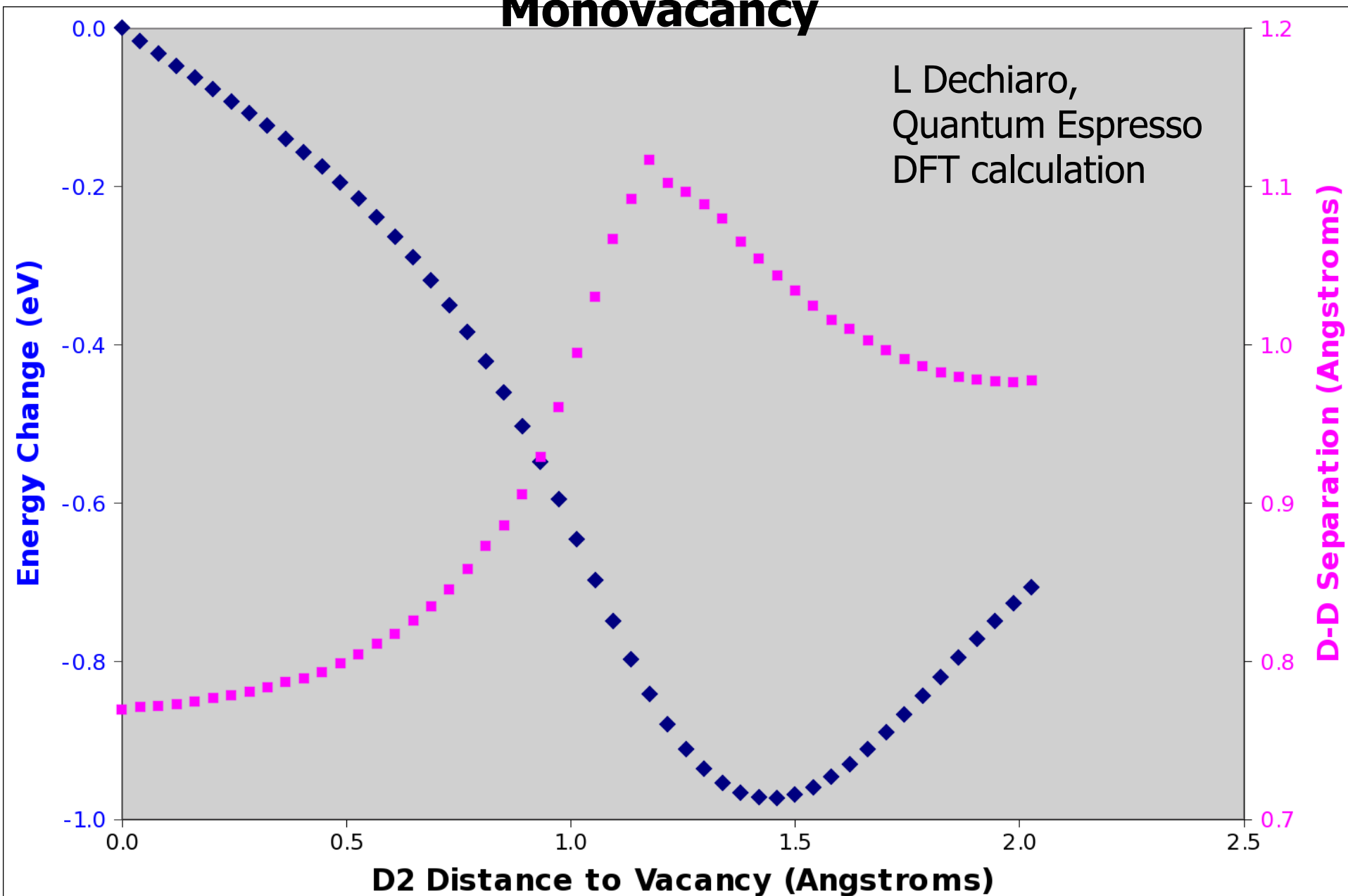
# Recall electron density near vacancy in Pd...



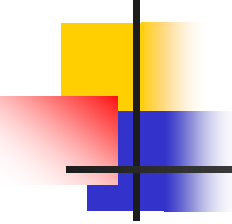
# [100] Displacement of D<sub>2</sub> in PdD Supercell

## Monovacancy

L Dechiaro,  
Quantum Espresso  
DFT calculation



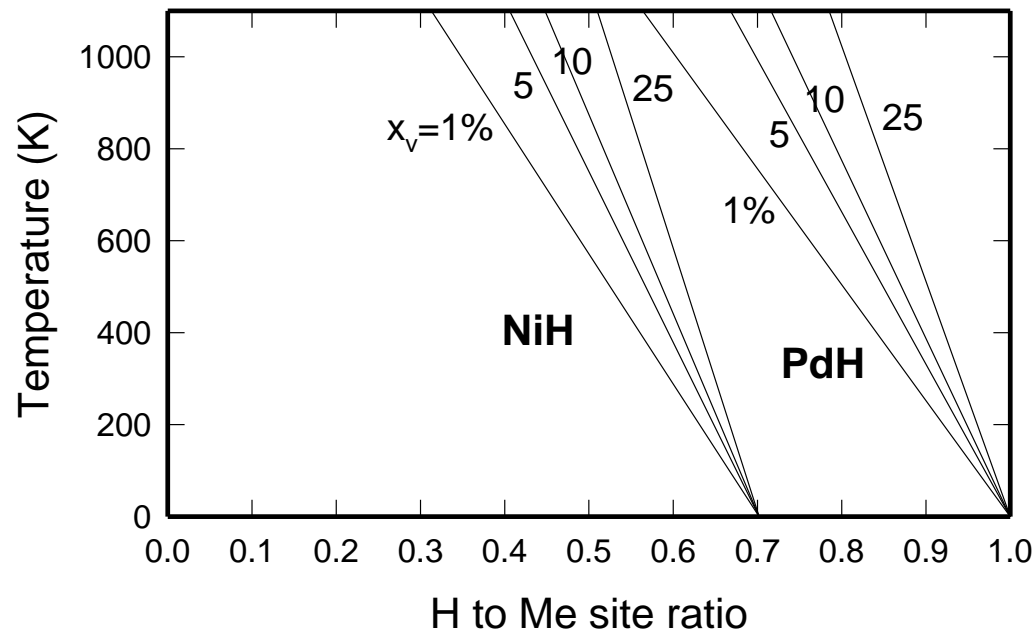




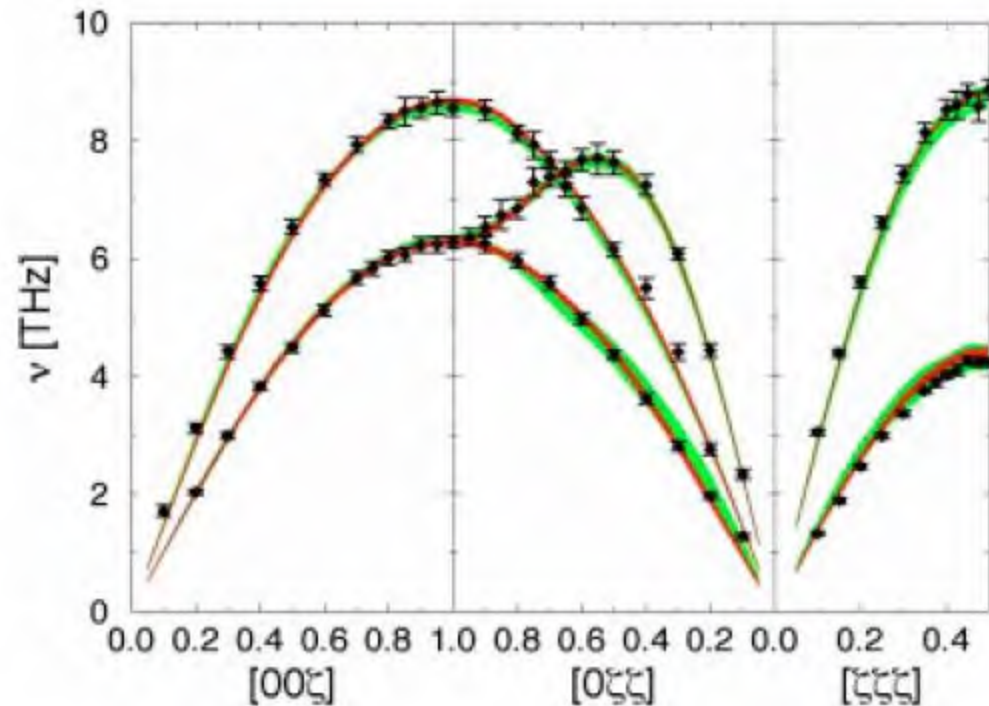
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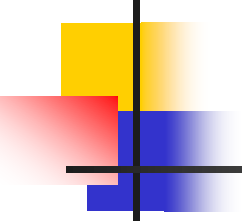
Electron density even higher in Ni (which is why H doesn't load well). But electron density reduced near a vacancy, and can form H<sub>2</sub>. Ni is closest analog of Pd for H<sub>2</sub>/D<sub>2</sub> formation near vacancy. Expect issues for vacancy creation and HD molecule formation to be similar.

# Vacancies made more readily



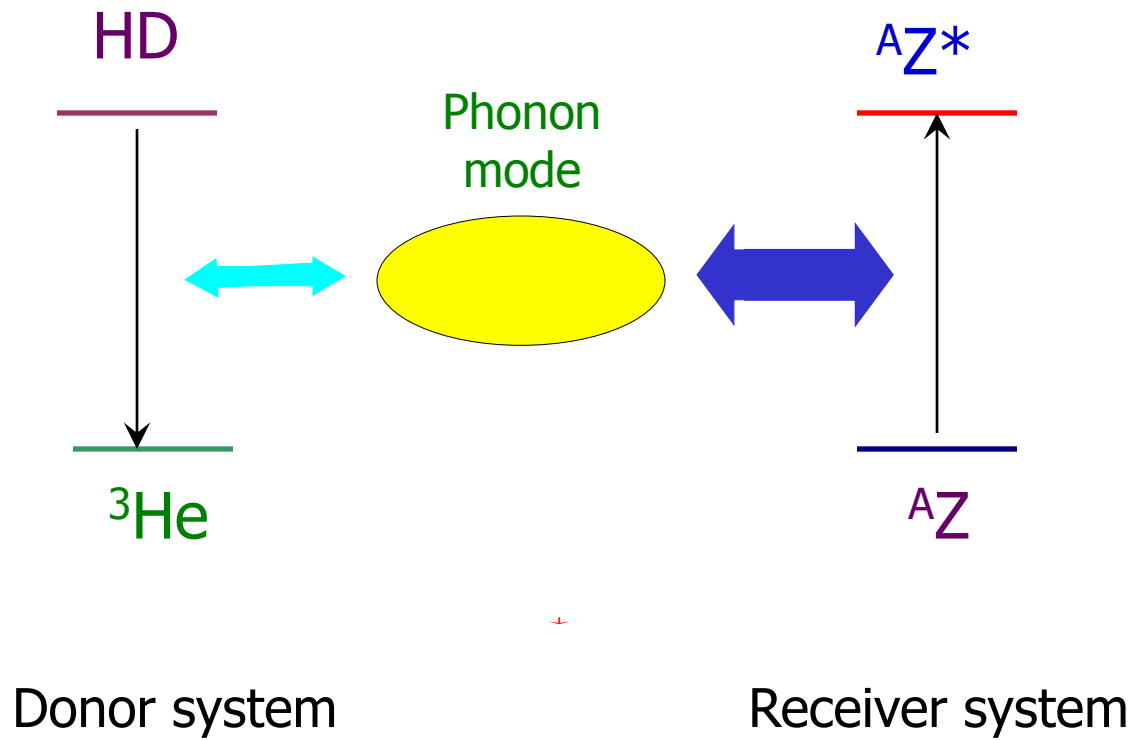
# Ni phonon modes



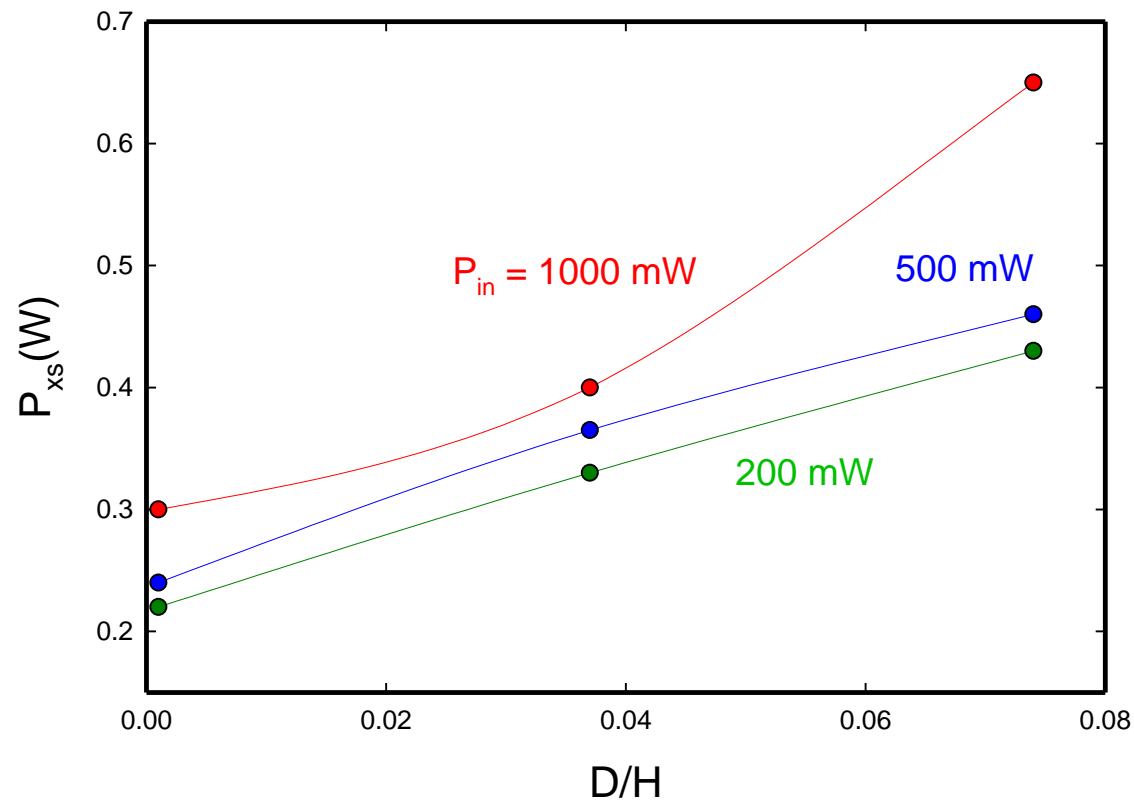


Issues with the development of optical phonon modes in gas loading since H concentration is low

# Donor-receiver model



# More $P_{xs}$ with D added





# More issues

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- HD/<sup>3</sup>He transition fine for donor
- Reduced mass smaller than for D<sub>2</sub>/<sup>4</sup>He system

$$\frac{1}{\mu} = \frac{1}{m_1} + \frac{1}{m_2} \quad \mu_{DD} = \frac{M_D}{2} = M_H \quad \mu_{HD} = \frac{2}{3}M_H$$

- So tunneling is orders of magnitude larger
- Deuterium natural abundance is 1/6240 of hydrogen
- $\Delta E = 5.49$  MeV, so need to exchange few quanta



# Take away message

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- Excess heat seen in NiH
- Electrochemical systems, gas systems
- Harder to load
- Easier to make vacancies
- HD formation good
- Donor-receiver model happy
- Is some D in H
- Larger interaction matrix element since Gamow factor smaller

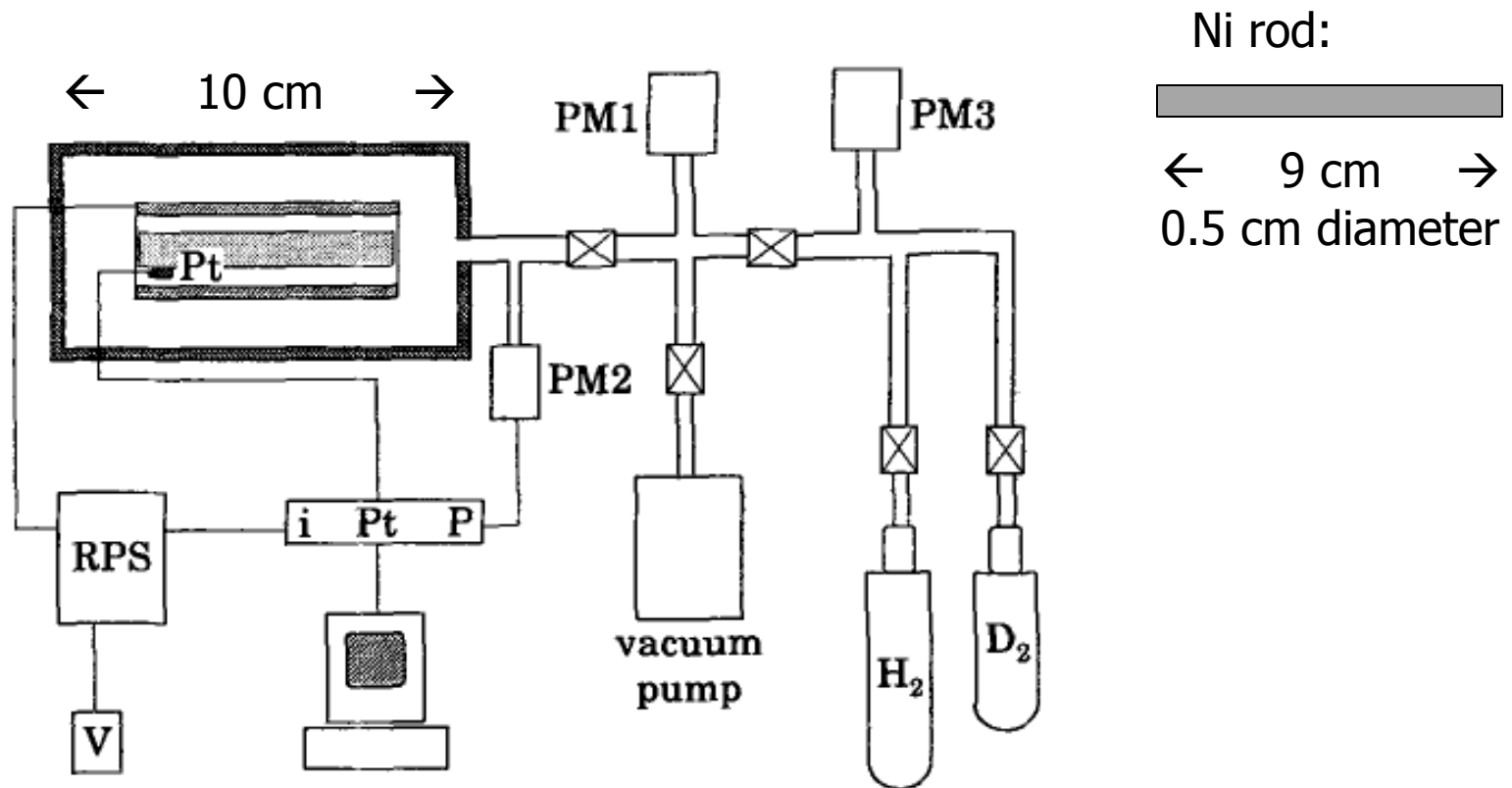




# Piantelli experiment

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# Piantelli experiment



# Calibration

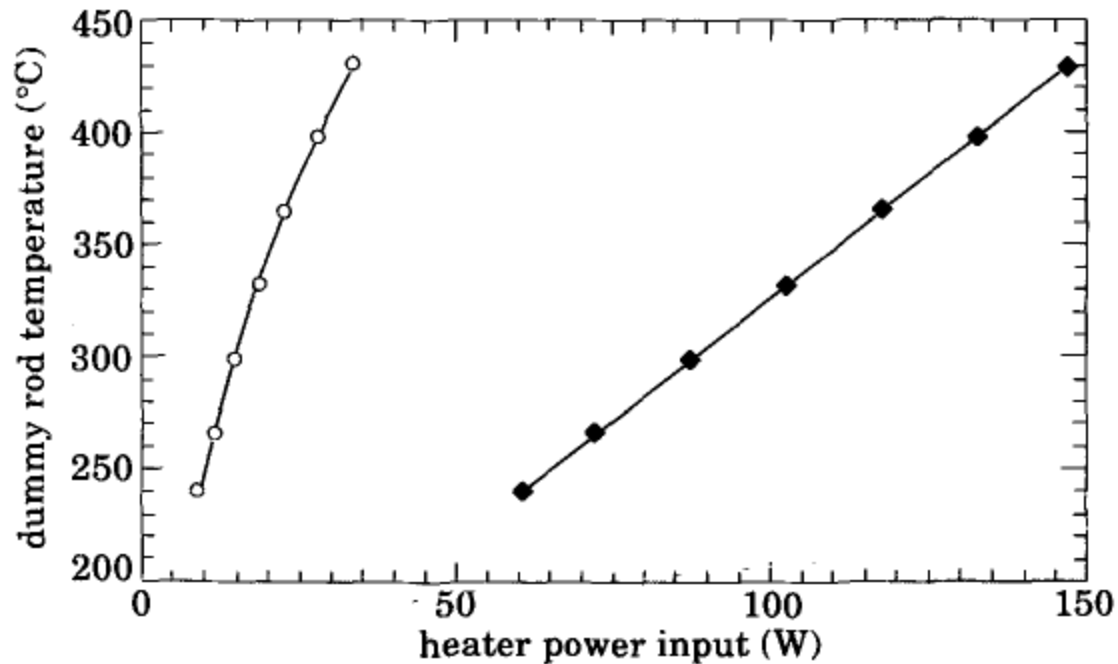
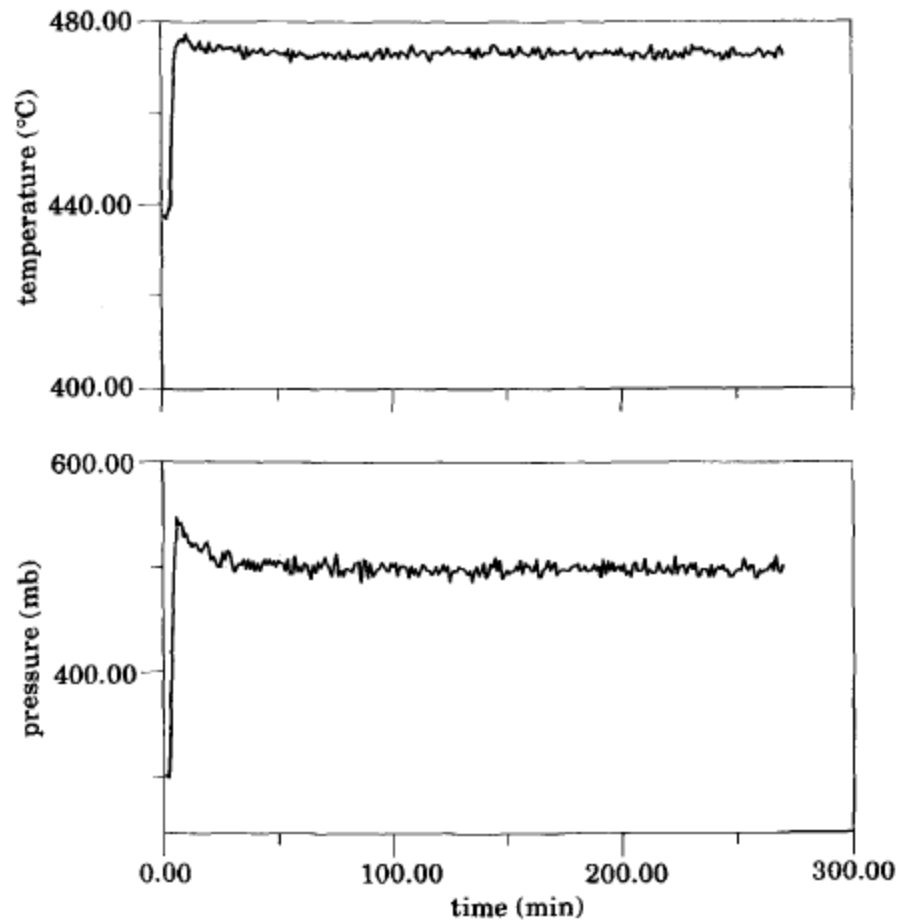


Fig. 2. – Power-temperature relations for the dummy rod at different values of the pressure inside the chamber. ◆ Hydrogen ( $p = 570$  mbar), ○ vacuum.

# Data showing $P_{xs}$



Input power: 140? W  
Excess power: 20 W

Fig. 3. – Sample temperature and hydrogen pressure vs. time, during an «anomalous» loading step.

# T vs $P_{in}$ for $P_{xs} = 0, 20, 50W$

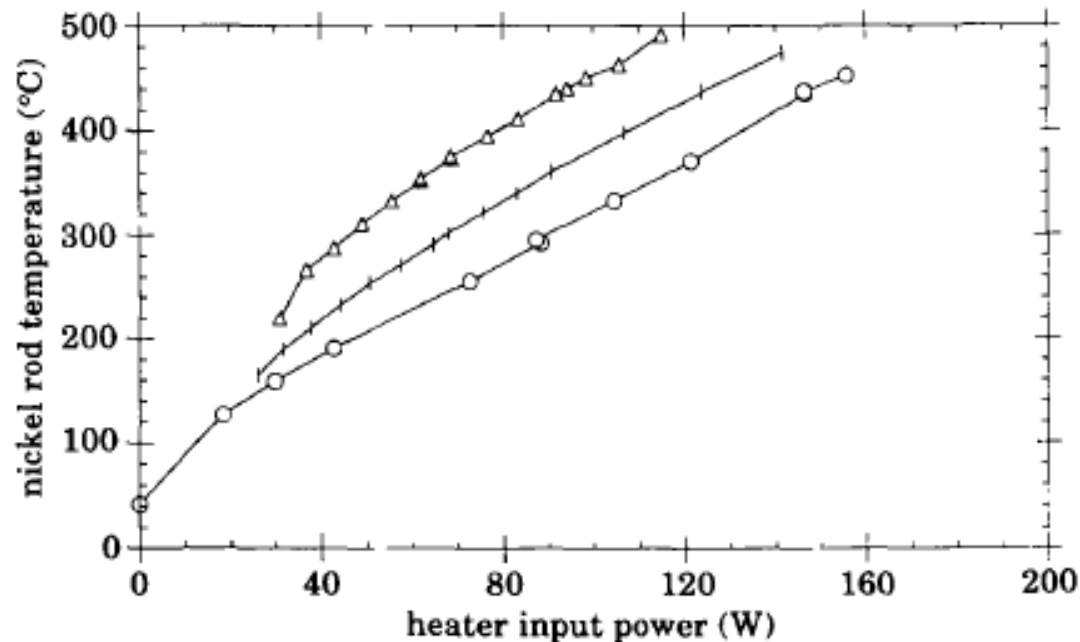


Fig. 4. - Temperature *vs.* heater-power curves family for a loaded nickel sample at different values of power «imbalance».  $\circ$  0 W,  $—+$  20 W,  $\triangle$  50 W. The dummy rod and the unloaded Ni rod are represented by the 0 W lower «imbalance» curve.

# 1998 Piantelli experiment

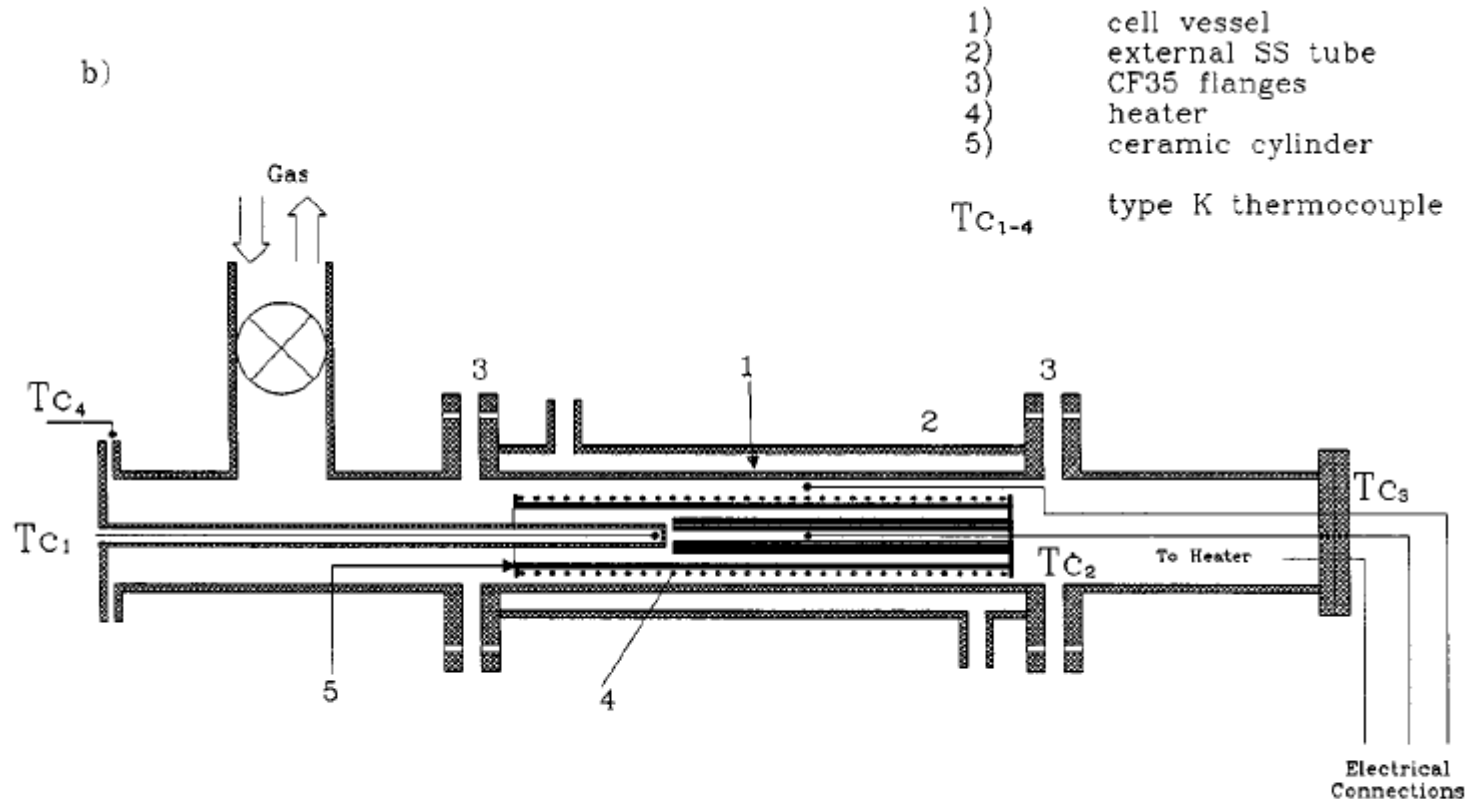


Fig. 1b. – Schematic drawing of cell B. Tc<sub>1</sub>, Tc<sub>2</sub>, Tc<sub>3</sub> and Tc<sub>4</sub> show the thermocouples positions.

S Focardi, V Gabbani, V Montalbano, F Piantelli, S Veronesi,  
*Il Nuovo Cimento* **111** 1233 (1998)

# Calibration curves

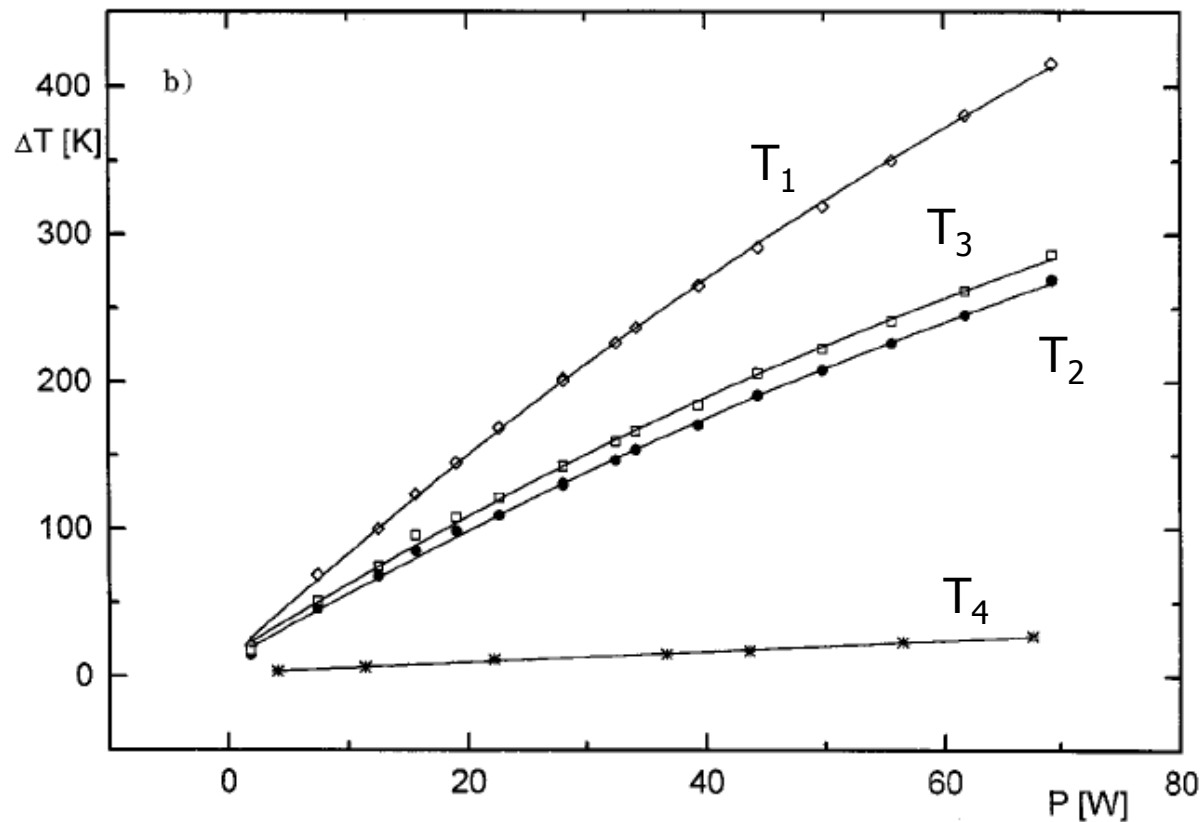
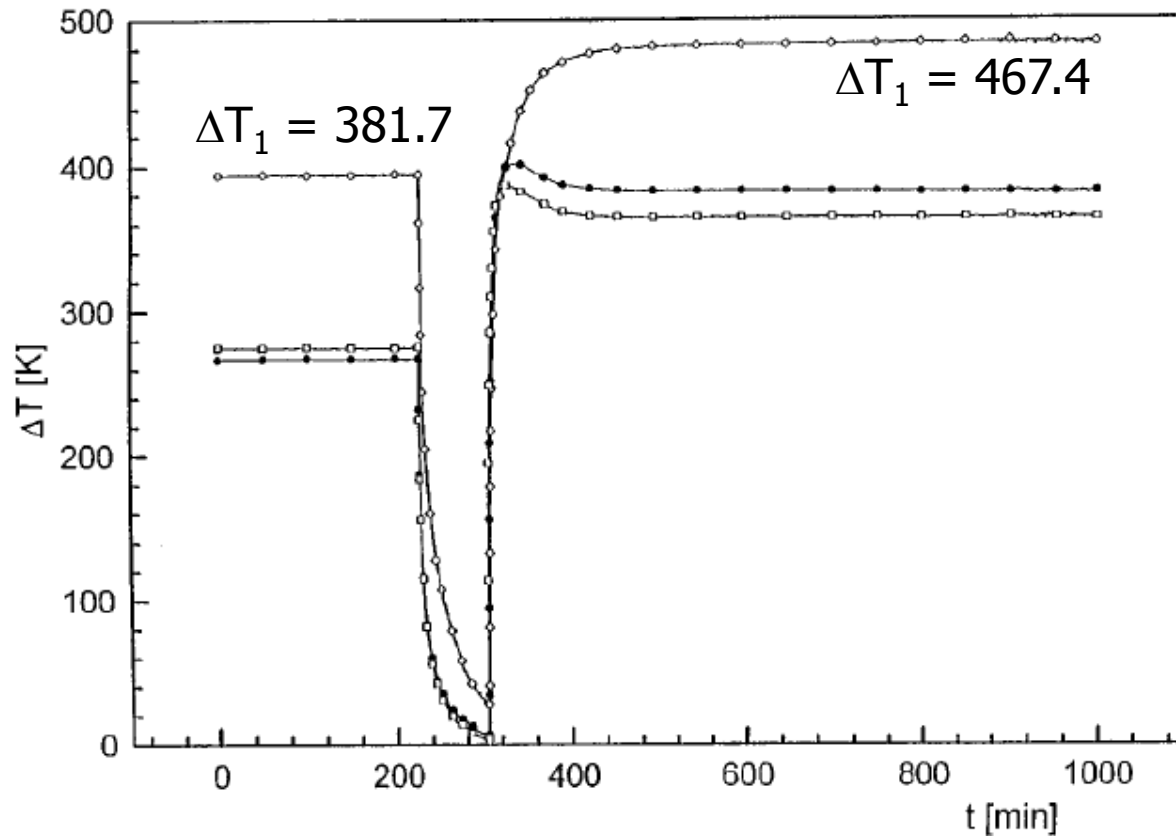


Fig. 2. – a) Cell A. Calibration curves; temperature (relative to the room temperature) *vs.* power ( $T_{Pt}(\diamond)$  and  $T_e(\bullet)$ ). b) Cell B. Calibration curves; temperature (relative to the room temperature) *vs.* power ( $T_1(\diamond)$ ,  $T_2(\bullet)$ ,  $T_3(\square)$  and  $T_4(*)$ ).

# Excitation of the sample



Approx:

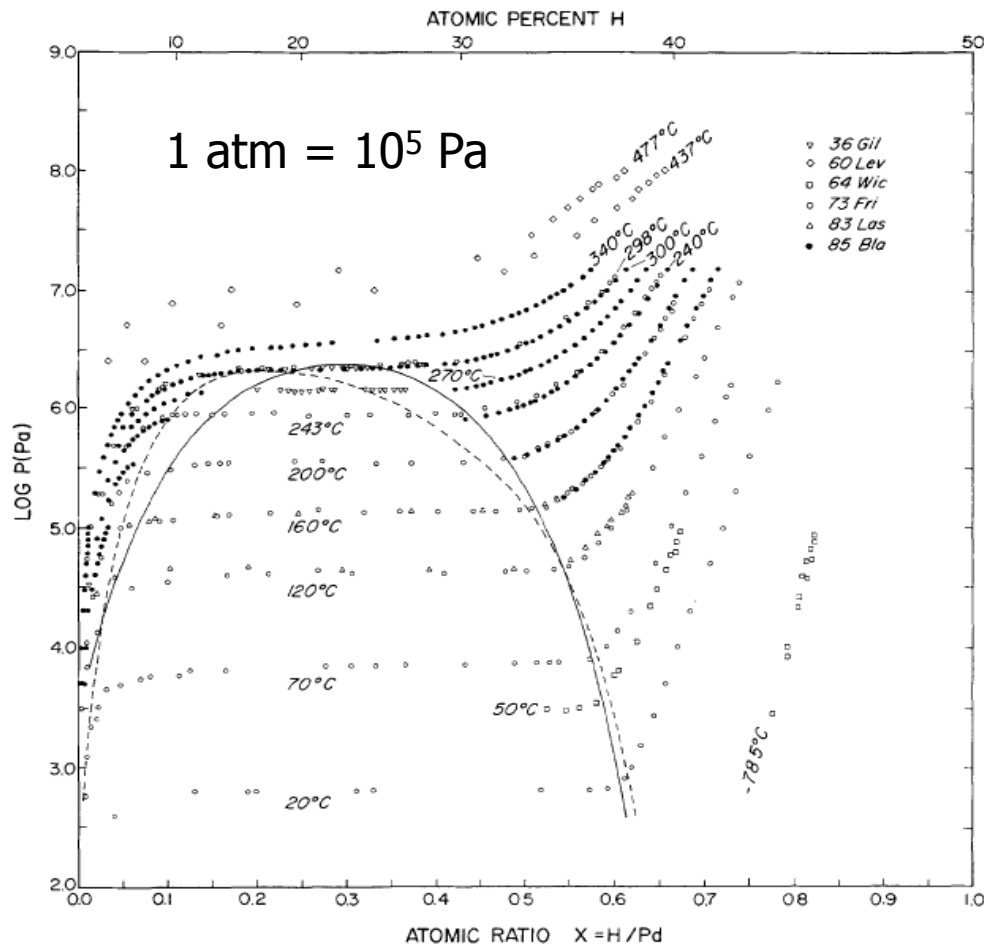
$$P_{in} = 60 \text{ W}$$

$$P_{xs} = 20 \text{ W}$$

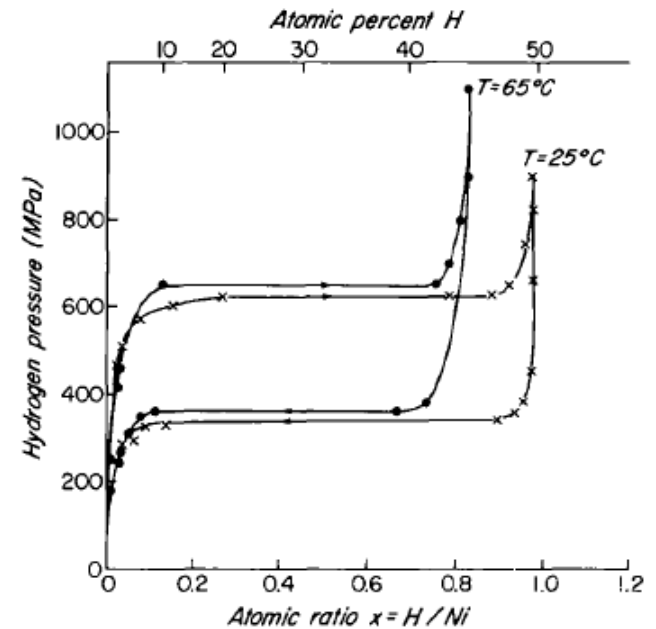
Fig. 4. – A sample excitation performed with a temperature jump for cell B ( $T_1$ ( $\diamond$ ),  $T_2$ ( $\bullet$ ),  $T_3$ ( $\square$ )). An inversion between  $T_2$  and  $T_3$  can be observed. Such an effect is due to the extra power produced by the nickel rod.



# Pressure-composition isotherms



**Fig. 4 Absorption and Desorption Isotherms at 25 and 65 °C for the Ni-H System**



From [78Bar]. M. Wayman and G.C. Weatherly, 1989.

# Solubility of H at low pressure

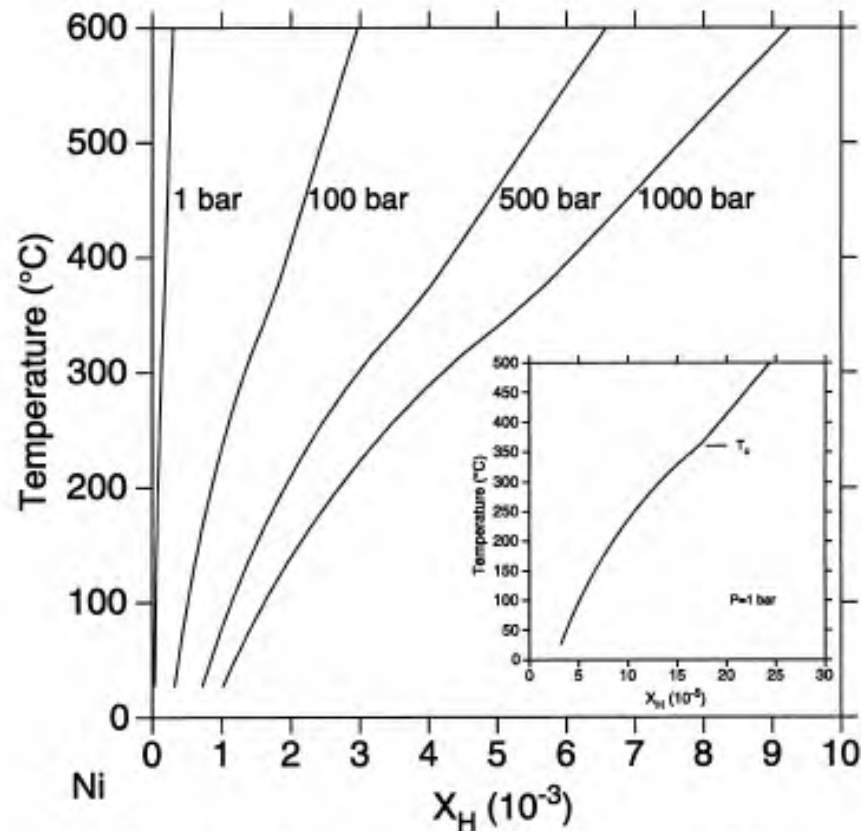


Fig. 7. Calculated solubility curves of hydrogen in nickel at different pressures, showing jogs at the Curie temperature. The insert is an enlarged diagram of the curve at 1 bar.

# Uptake of $\text{H}_2$ after several loading cycles

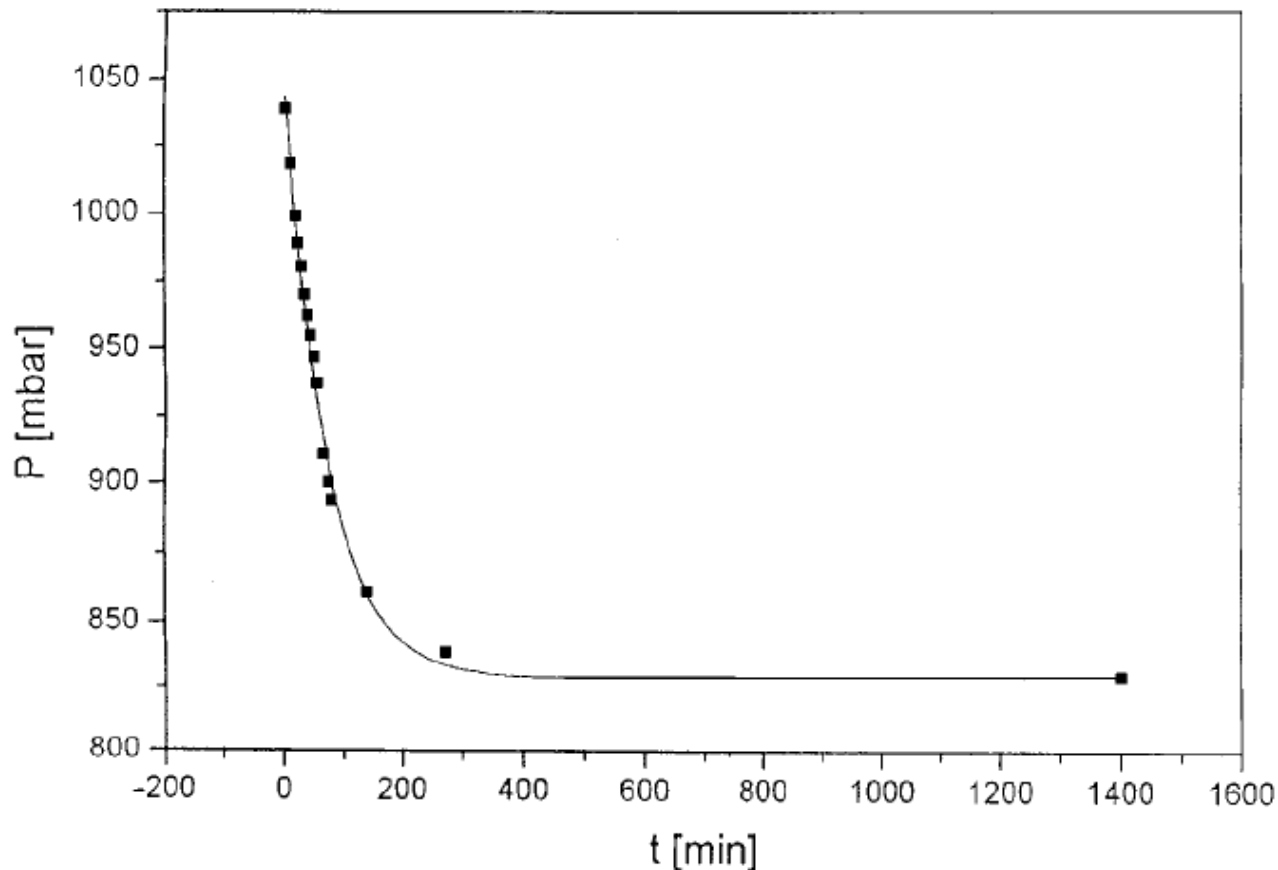


Fig. 3. – A typical hydrogen loading of a sample. The plot refers to cell B. The experimental data can be fitted with an exponential law. The characteristic time for this data set is  $\tau = 72$  min.



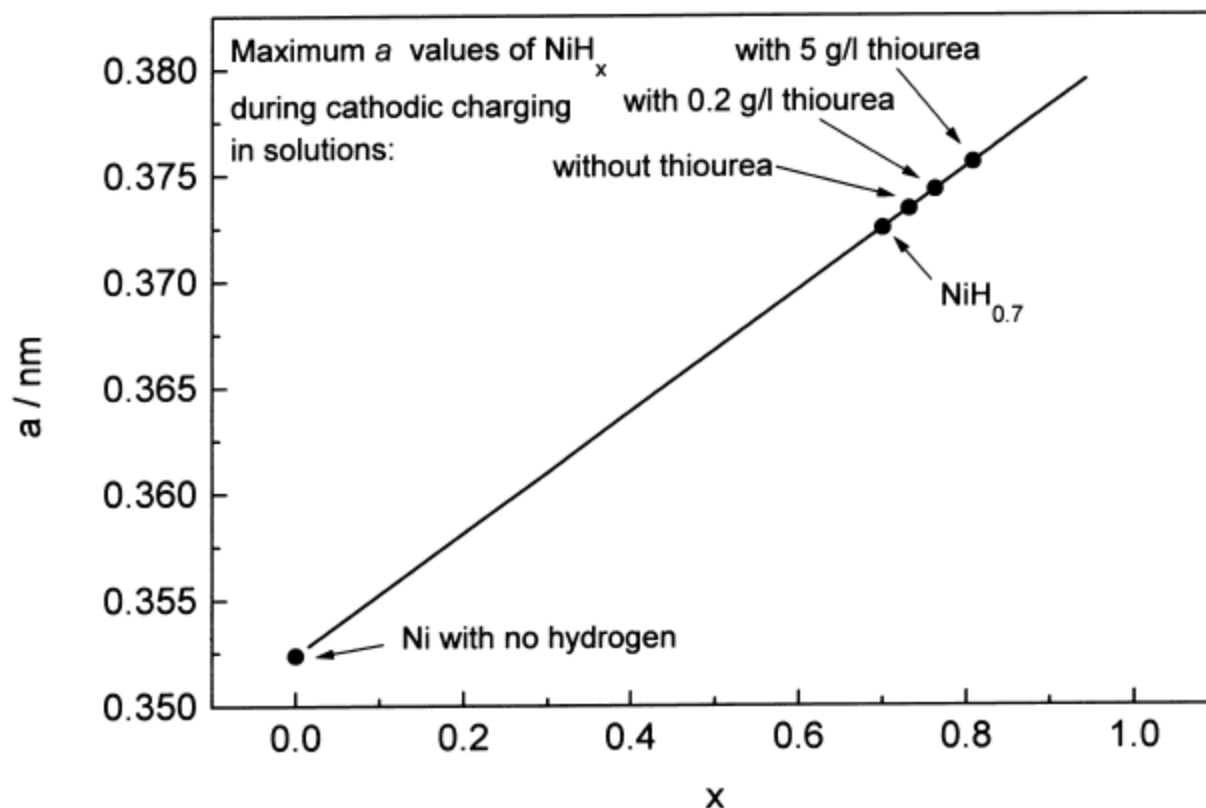
# Thinking about result

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- Bulk Ni does not load much ( $5\text{-}20 \times 10^{-5}$ ) near 1 atm
- Need  $O(6000 \text{ atm})$  to pressure-load bulk Ni with  $\text{H}_2$
- But some loading observed nonetheless in Piantelli expts
- Number of H atoms absorbed is several times  $O(3 \times 10^{21})$
- Number of Ni atoms in sample (1994) is  $O(4 \times 10^{23})$
- Loading in Cammarota replication from  $\text{H}_2$  absorption is  $\text{NiH}_{0.2}$
- Must be (non-bulk) special sites (defects or impurities)
- But not enough impurities in Cammarota version!

# X-ray diffraction data in electrochemical loading

Juskenas et al, *Electrochimica Acta* **43** 1903 (1998)



No evidence for intermediate  $a$  values for loadings in the  $\alpha$ - $\beta$  phase region. Observed only is the change in volume occupied by  $\beta$ -phase  $\text{NiH}$ .

Fig. 8. Dependence of calculated (solid line) and experimental (●) values of the lattice parameter  $a$  of  $\beta$ - $\text{NiH}_x$  phase on the stoichiometric coefficient  $x$  of the hydride phase.

# Diffusion of H

- Diffusion of H in Ni is much slower than in Pd

$$D = D_0 e^{-\Delta E/kT}$$

$$D_0 = 7.04 \times 10^{-3} \text{ cm}^2/\text{sec}$$

$$\Delta E = 409 \text{ meV}$$

$$D(300\text{K}) = 9.5 \times 10^{-10} \text{ cm}^2/\text{sec (NiH)}$$

$$D(300\text{K}) = 5.5 \times 10^{-7} \text{ cm}^2/\text{sec (PdD)}$$

- Elevated temperature D in NiH is similar to D in PdD at 300 K

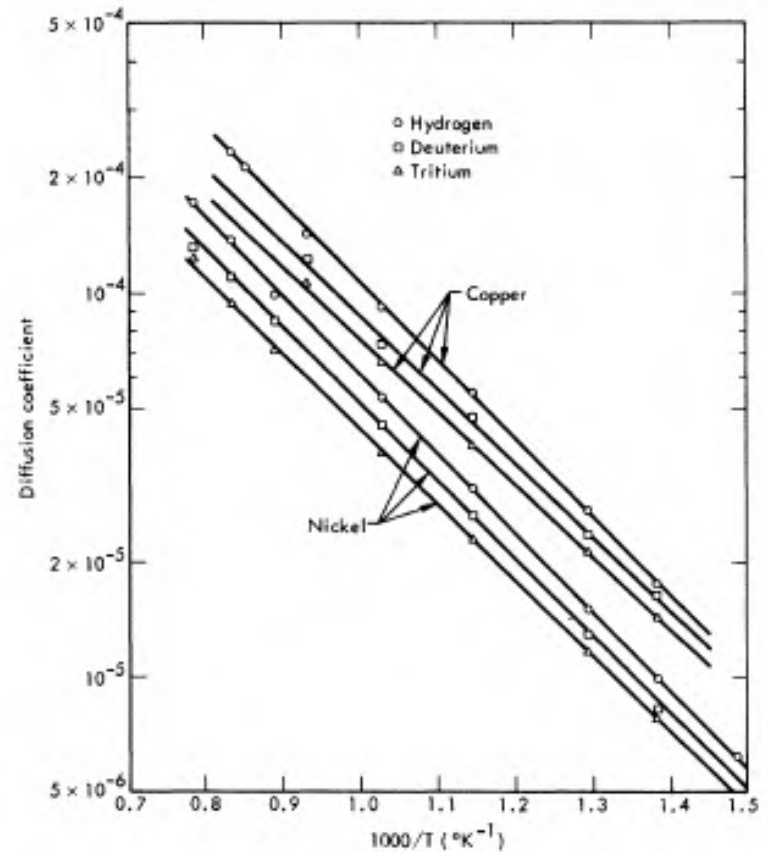
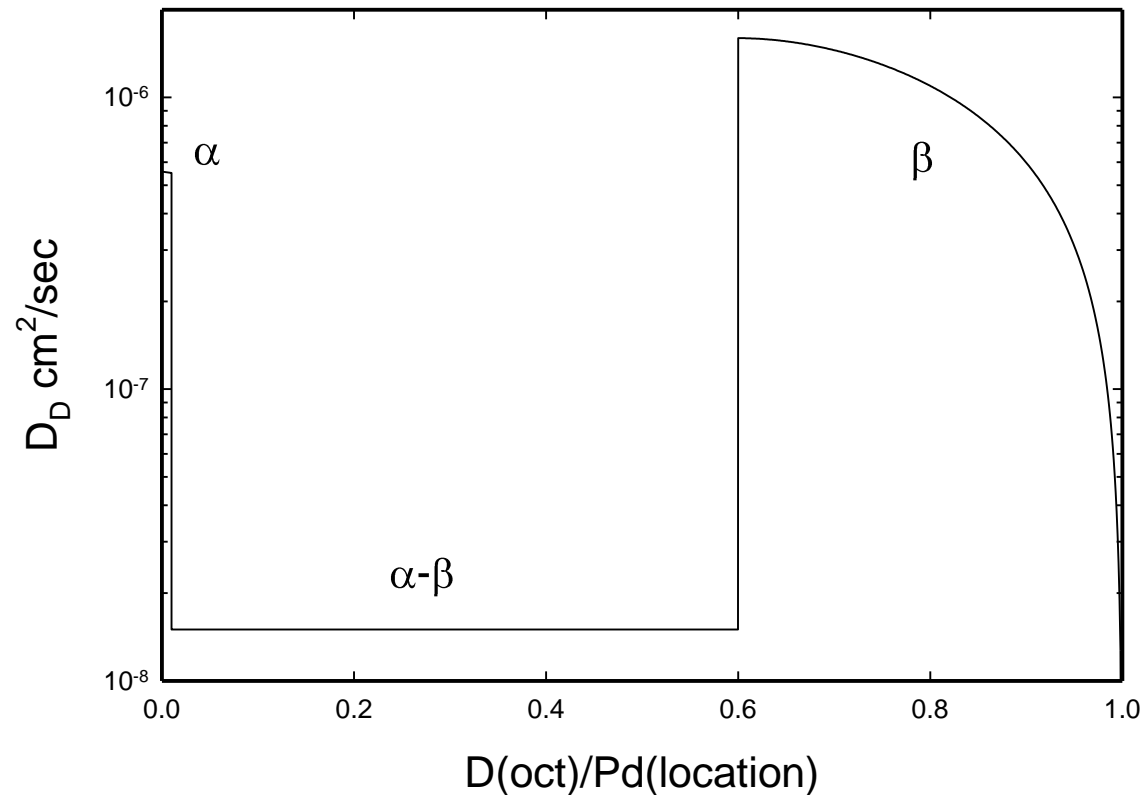
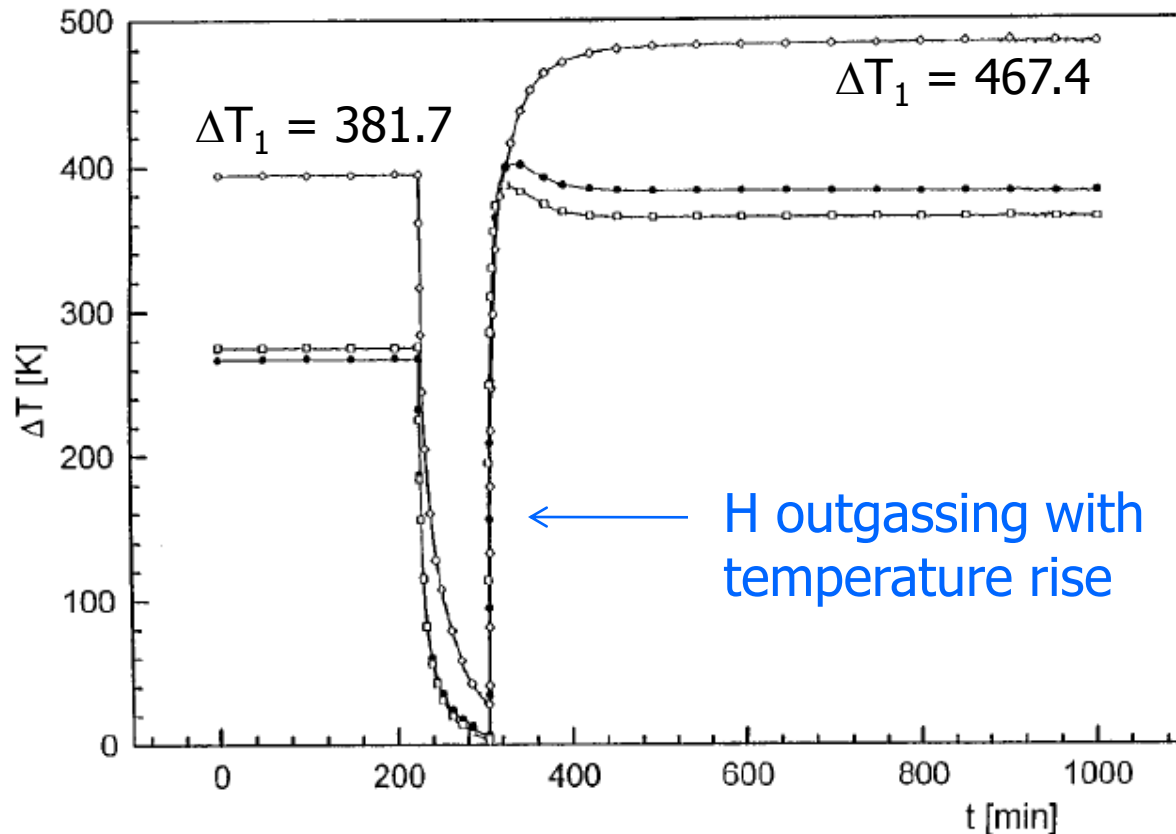


FIG. 3. Arrhenius plots for H<sub>2</sub>, D<sub>2</sub>, and T<sub>2</sub> diffusing in Cu (upper graphs) and Ni (lower graphs).

# PdD diffusion model at 300 K



# Excitation of the sample



Approx:

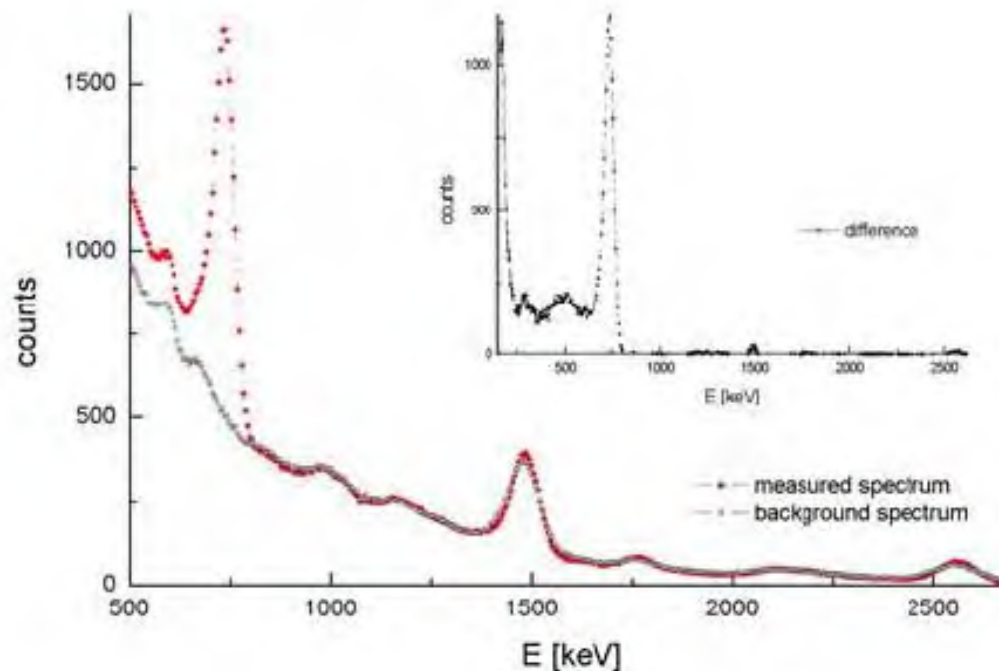
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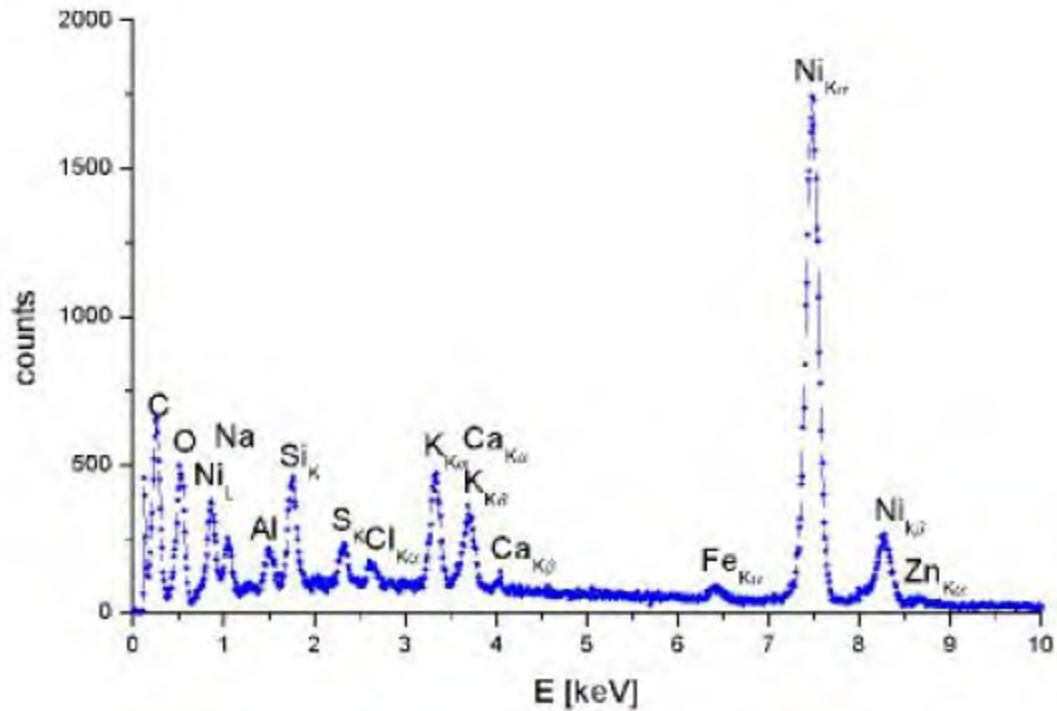


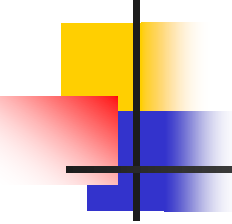
# $\gamma$ -emission events



S Focardi and F Piantelli, "Produzione de energia e reazioni nucleari in sistemi NiH a 400 C" (2000)

# New elements in Piantelli expt



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- 
- Low H loading in NiH gas systems
  - Not enough H for good optical phonon mode
  - NiH systems so far probably work based on acoustic mode excitation
  - Ni then participates strongly in vibrations
  - Coupling of energy through nuclear excited states in Ni
  - Some long-lived ones will have fission decay pathways
  - Lattice-induced fission produces new elements
  - Eats up significant amount of produced energy