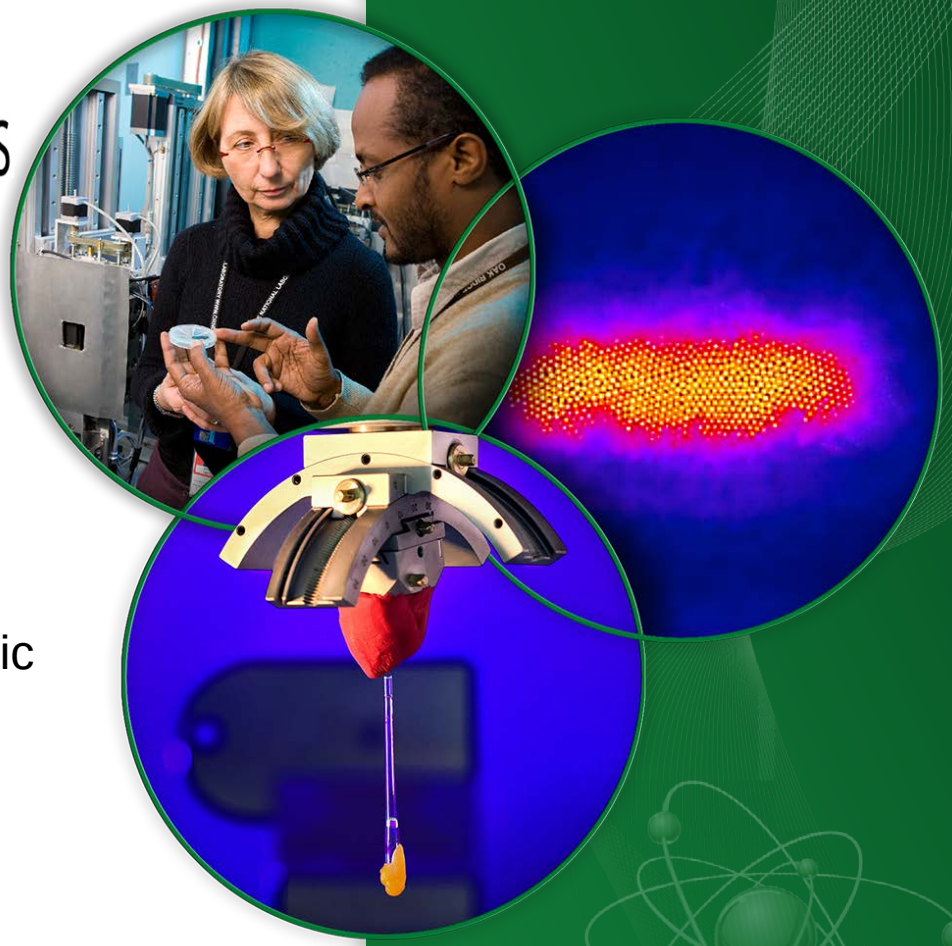
Neutron and X-ray School 2016
Oak Ridge/Argonne

ARCS Experiment
August 1-4, 2016



Dynamics of metal hydride systems: Harmonic oscillators and beyond

Or

Is ZrH2 a collection of quantum harmonic oscillators?

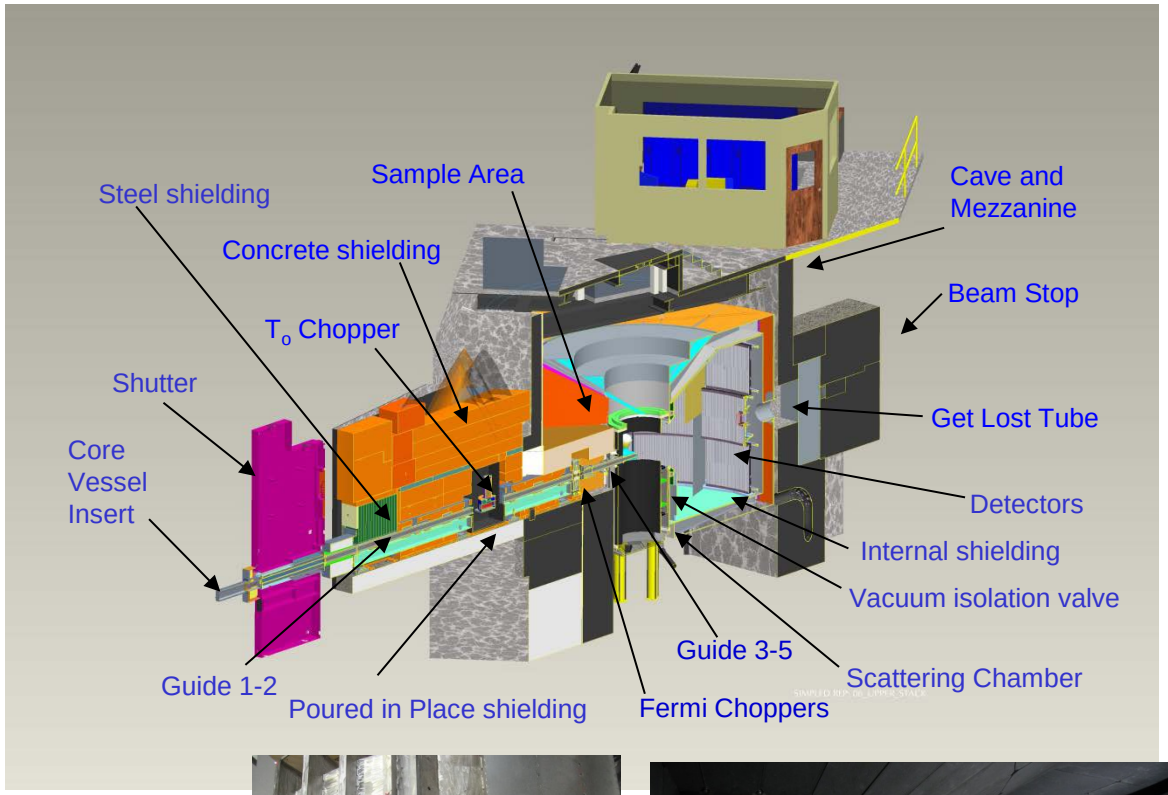
Doug Abernathy, Andy Christianson, Lacy Jones, Victor Fanelli

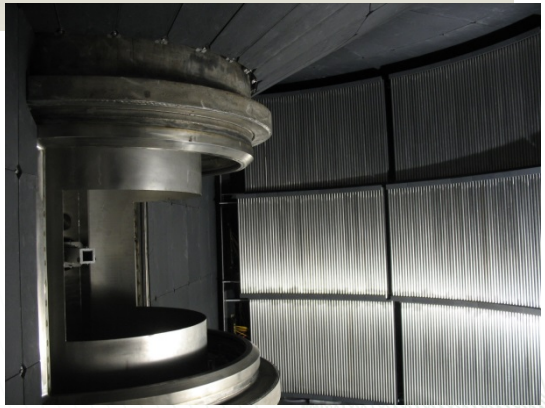
ORNL is managed by UT-Battelle
for the US Department of Energy

ARCS overview - instrument

SPECIFICATIONS

Moderator	Decoupled ambient water
Source-to-Fermi chopper distance	11.6 m
Chopper-to-sample distance	2.0 m
Sample-to-detector distance	3.0 to 3.4 m cylindrical geometry
Incident energy range	10–1500 meV
Resolution (elastic)	2–5% E_i
Detector coverage horizontal	-28–135°
Detector coverage vertical	-27–26°
Minimum detector angle	3°





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ARCS overview - applications

- Lattice Dynamics
  - Entropy and the effects of vibrational modes on stability and phase transitions of solids
  - Excitations in disordered materials; effects of nanoscale features on vibrational entropy and thermodynamic stability
  - Equations-of-state from the measured phonon density-of-states versus temperature and pressure
  - Phonons in correlated-electron materials; coupling of lattice and electronic degrees of freedom in high-T_c, heavy-fermion, and mixed-valence materials
- Magnetic Dynamics
  - High-temperature superconductivity—spin dynamics in superconductors and precursor compounds and crystal field spectroscopy
  - Low-dimensional systems; one-dimensional quantum magnets and low-dimensional conductors
  - Magnetism in actinide materials; heavy-fermion magnetism and superconductivity
- Chemical Physics
  - Deep inelastic neutron scattering studies of hydrogen, helium

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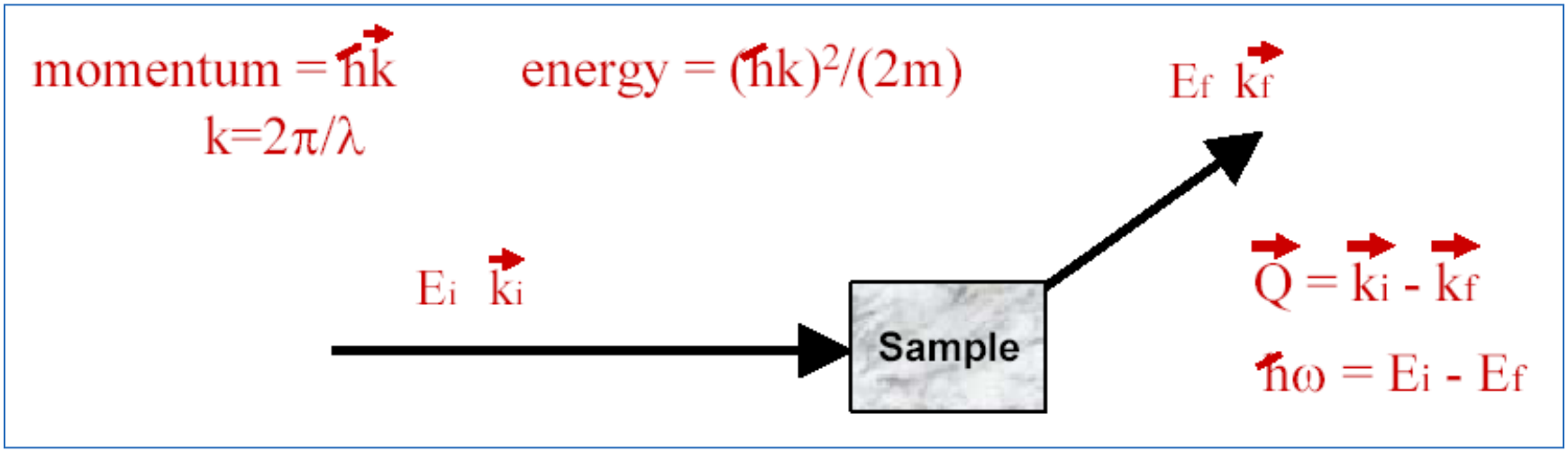
Inelastic neutron scattering geometry



Measure the number of scattered neutrons as a function of Q and ω

=> S(Q,ω) (the scattering function)

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k_f}{k_i} S(Q,\omega)$$

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Spectroscopy instrument types

- Direct geometry
  - E_i fixed
  - ARCS, SEQUOIA, CNCS, HYSPEC
- Inverse geometry
  - E_f fixed
  - BASIS, Vision
- Triple Axis
- Spin Echo

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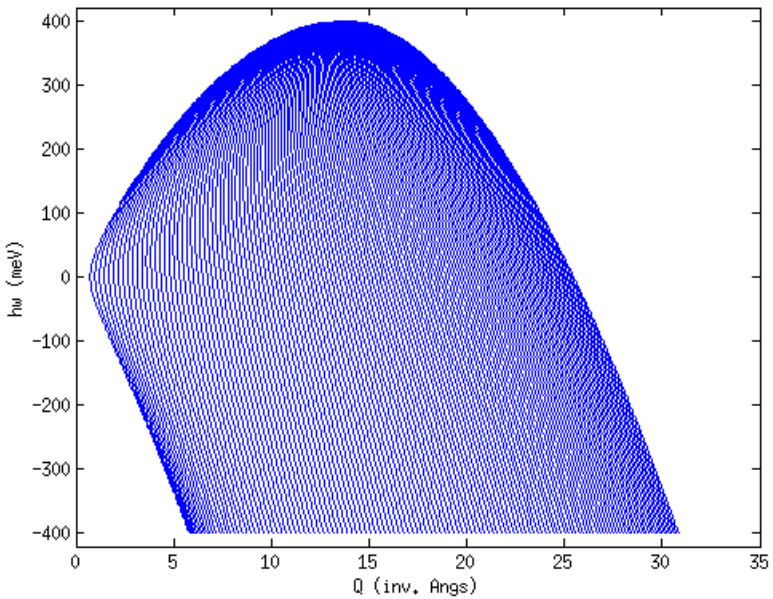
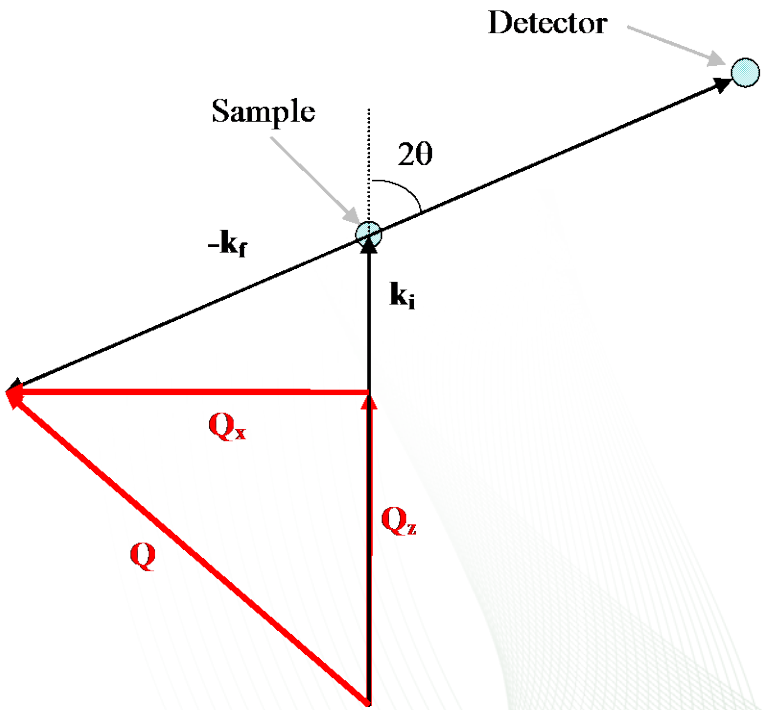
Direct geometry inelastic instruments



- Range of |Q| and ω is set by the detector coverage (Φ = 2θ) and incident energy E_i



Conservation Laws

$\hbar\omega = E_i - E_f$ Energy

$\vec{Q} = \vec{k}_i - \vec{k}_f$ Momentum

$$E = \frac{1}{2}mv^2 = \frac{p^2}{2m} = \frac{\hbar^2 k^2}{2m}$$

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Fermi chopper spectrometer – time/distance diagram

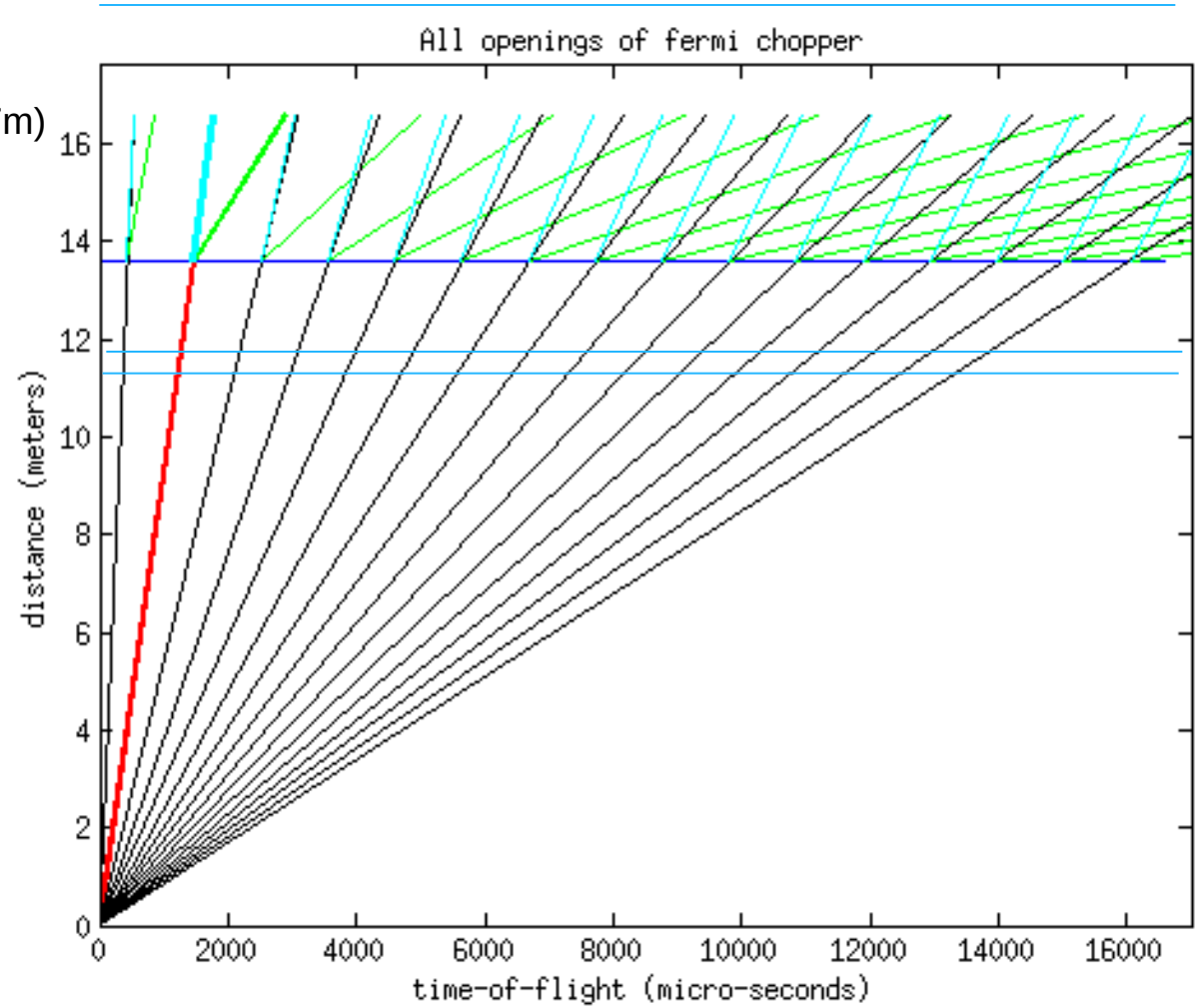
Monitor 2 (18.5m)

Detectors (16.6 to 17m)

Sample (13.6m)

Monitor 1 (11.831m)

Fermi (11.61m)



- **Fermi chopper spins at a multiple of the source frequency (higher gives better energy resolution but more overlap)**
- **Example for all energies transmitted**

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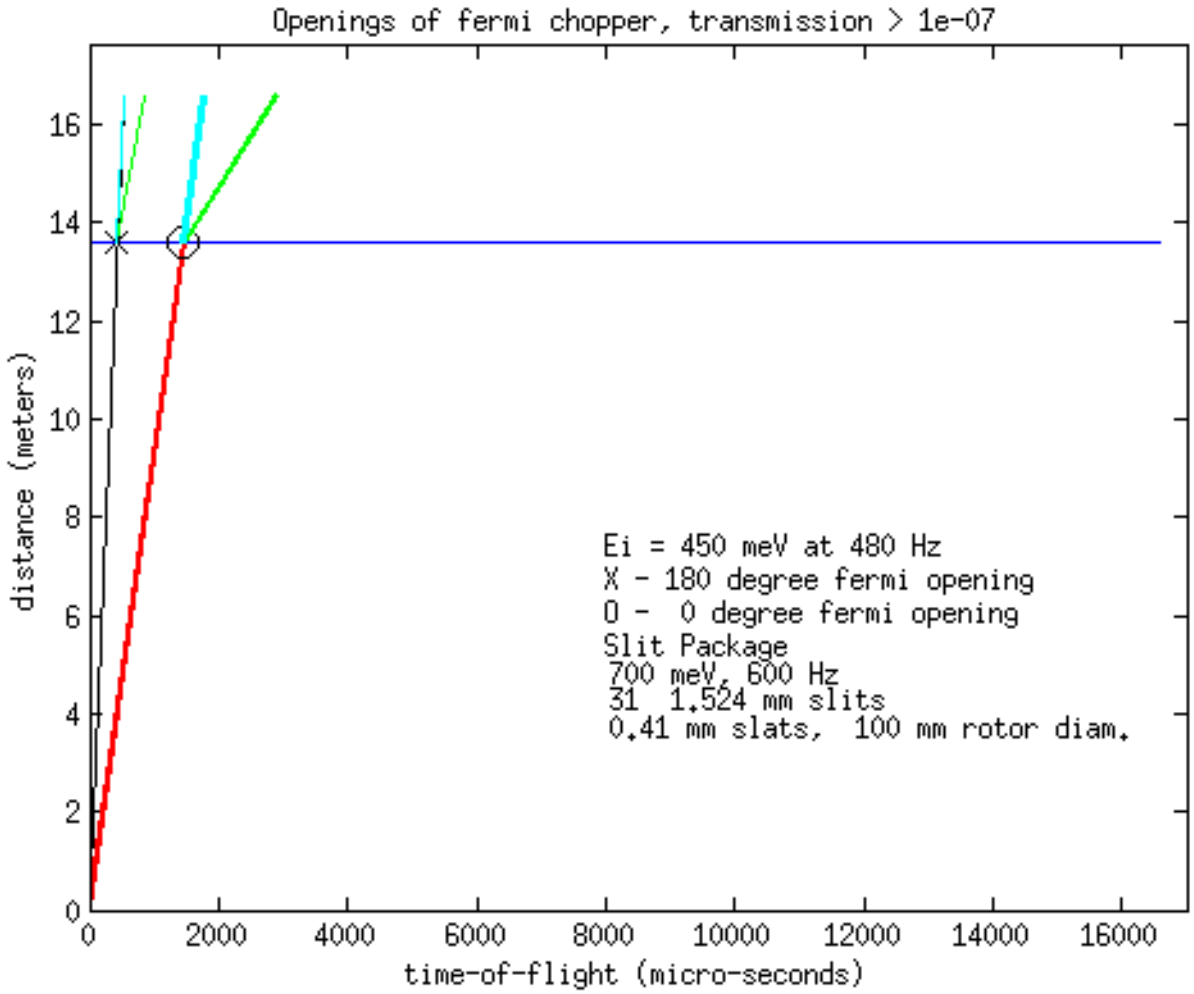
Fermi chopper spectrometer – time/distance diagram

- Fermi chopper transmission function applied
- Transmission limited due to mismatch of absorbing slat curvature with neutron velocity.
- Vertical axis T-zero chopper also cuts unwanted openings



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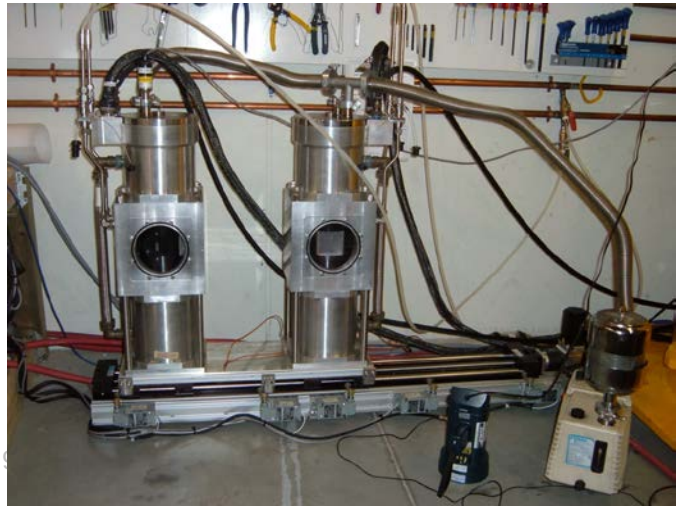
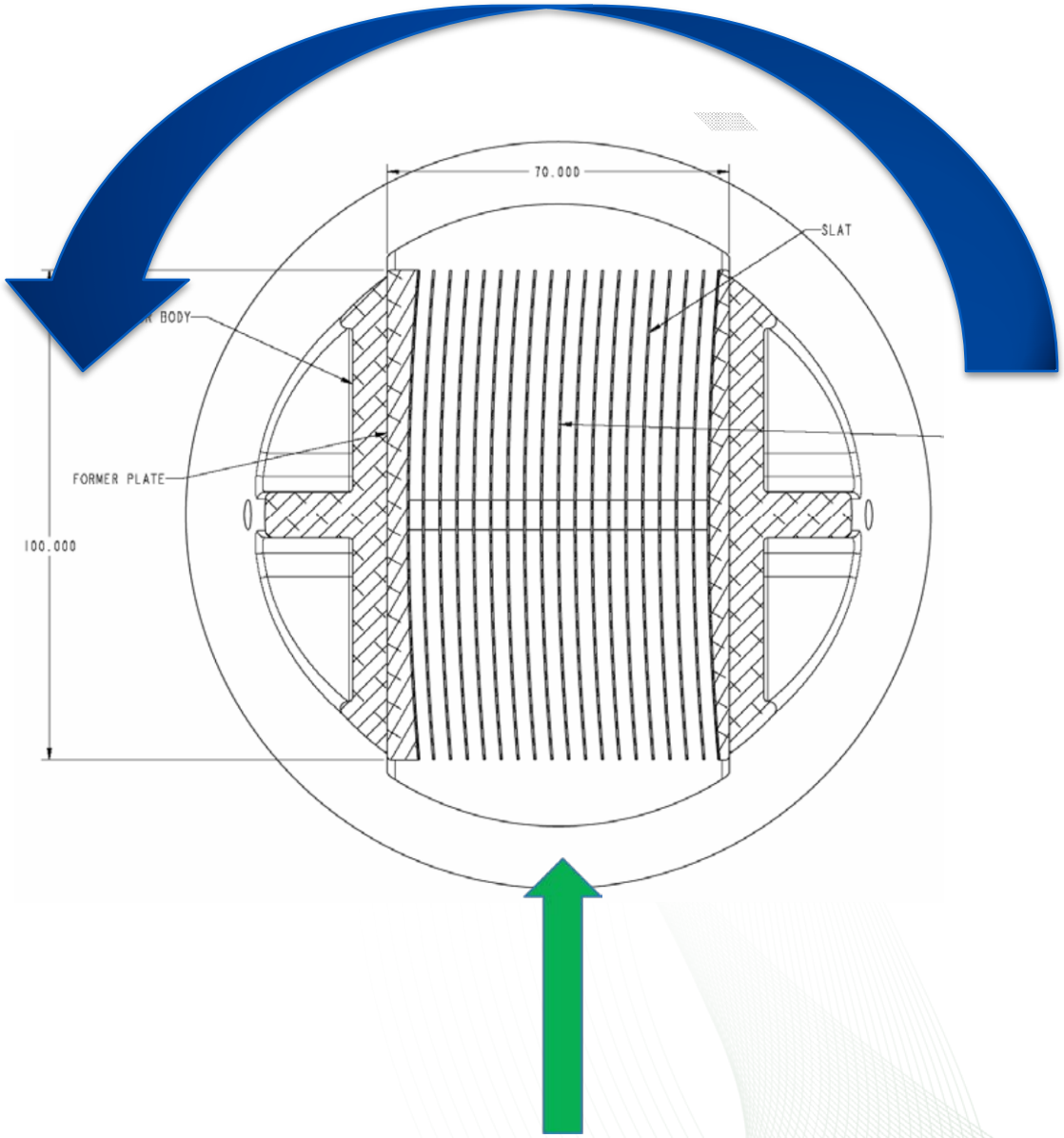
Fermi choppers

- Phase controls E_i (15 - 1500 meV)
- Frequency controls resolution; 0-600 Hz in increments of 60Hz.
- Magnetic bearing
- Two on a translation table
  - ~4% energy resolution (100meV range)
  - ~4-8% energy resolution (700meV range)





Vertical-axis T_0 chopper

- Based on IPNS chopper concept
- Procured with SEQUOIA and HYSPEC from SKF Magnetic Bearing
- Specifications:
  - Maximum speed 180Hz, steps of 30Hz
  - Minimum of 20cm Inconel in beam
  - Magnetic bearings for low maintenance
  - Remote handling capability

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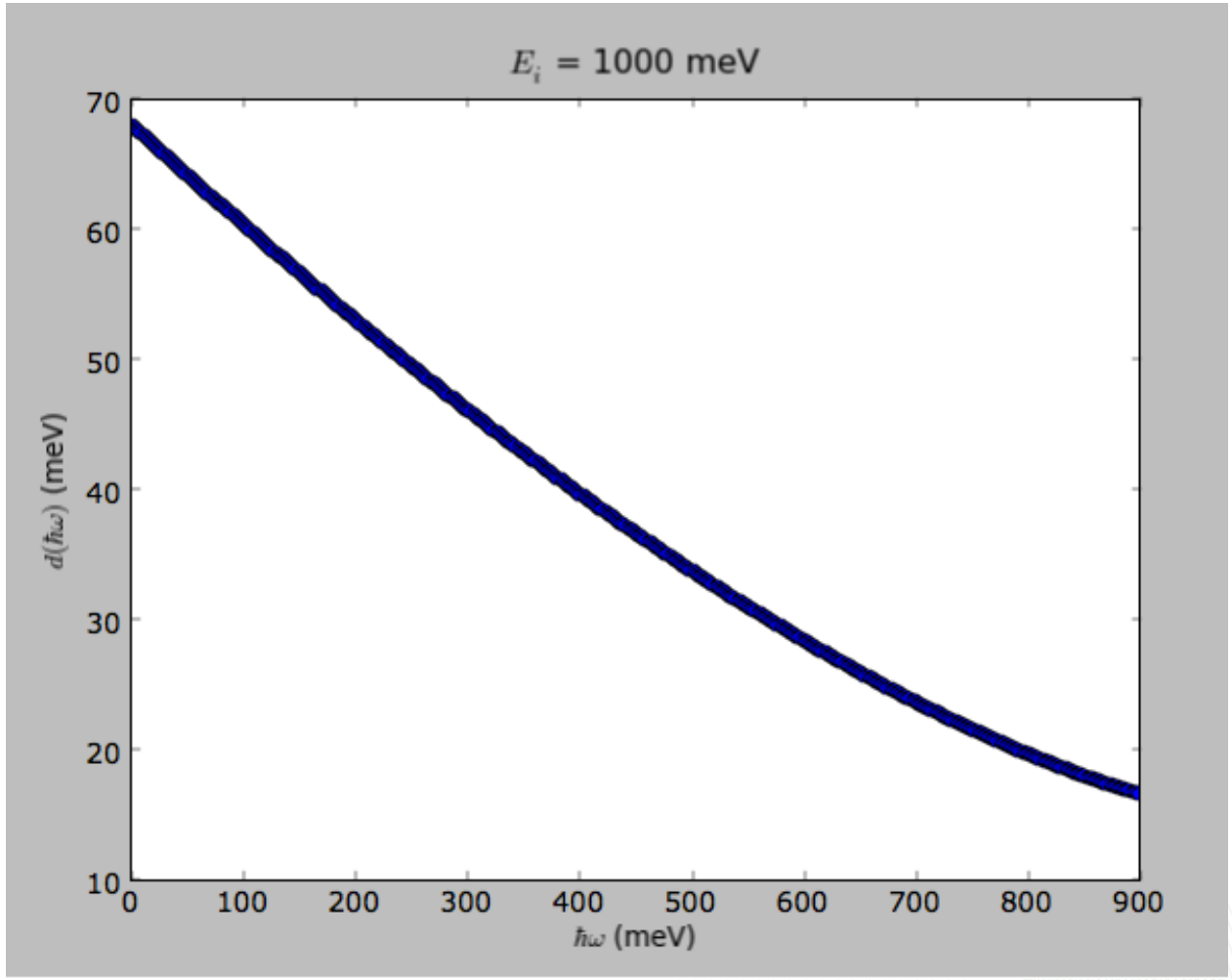
Energy resolution as a function of energy transfer

- Consequence of time-of-flight technique for direct geometry instruments:
  - Positive energy transfer ⇨
  - slower scattered neutrons ⇨
  - longer time-of-flight ⇨
  - better energy resolution



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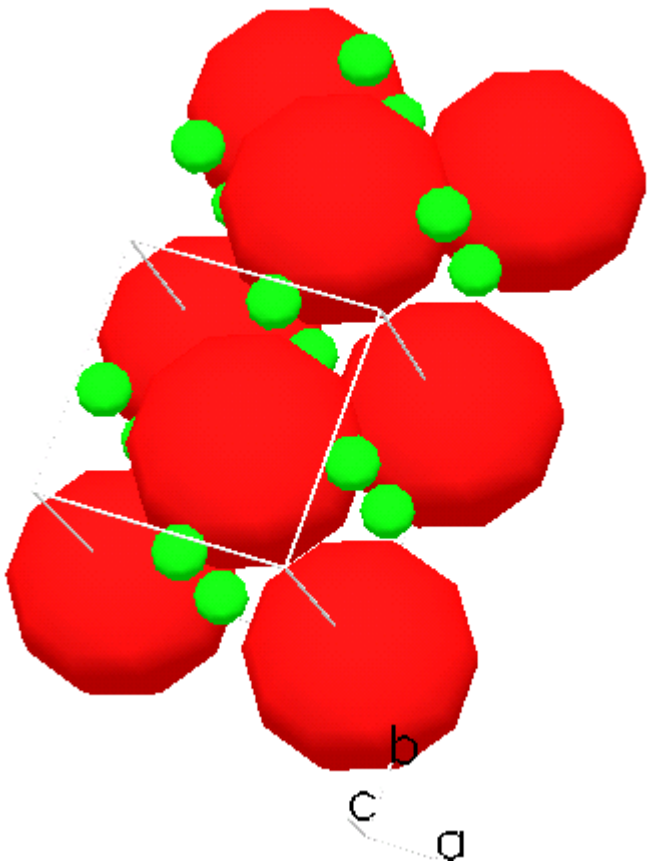
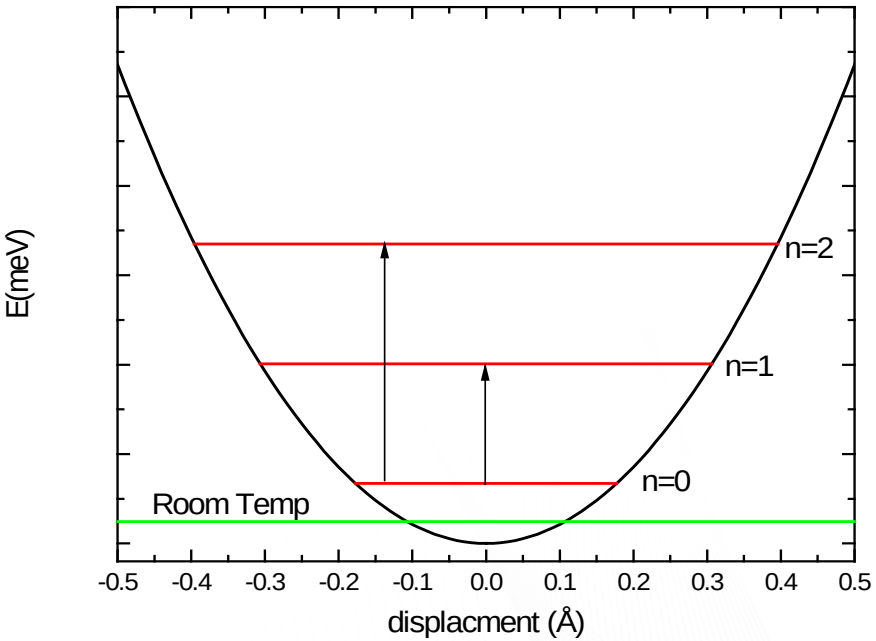
Is the H in ZrH_2 a quantum harmonic oscillator?



$E_n = (n+1/2)\hbar\omega \quad \hbar\omega = ? \quad n = 0, 1, 2, \ldots$



- Hydrogen in a potential well

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Predictions for a quantum harmonic oscillator

- **Excitation levels are evenly spaced and single mode:**
  - $E_n = (n+1/2)\hbar\omega \quad \hbar\omega = ? \quad n = 0, 1, 2, \ldots$
- **Intensity as a function of momentum transfer Q for elastic scattering follows a Debye-Waller form (rms displacement):**
  - $I(Q,E=0) = A \exp(-\langle u^2\rangle Q^2) + C$
- **The excitation energy and rms displacement should be consistent**
- **Intensity as a function of Q for the first excitation should follow:**
  - $I(Q, E=\hbar\omega) = B\, Q^2 \exp(-\langle u^2\rangle Q^2) + D$
- **Higher excitations should have equivalent forms with a term**
  - $I(Q,n\hbar\omega) \sim Q^{2n} \exp(-\langle u^2\rangle Q^2)$

How does our experiment match up? What could be causes of discrepancies?

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Possible additional measurements

- **Change incident energy**
  - **How does resolution change?**
  - **What other information about the system can be seen?**
- **Additional measurements to see instrumental effects**
  - **Measure empty can for background subtraction**
  - **Measure without collimator**
- **Different system: Uranium Nitride**
  - **QHO scattering seen while trying other experiment on the magnetism in UN**
  - **Reference: A.A. Aczel, G.E. Granroth, G.J. MacDougall, W.J.L. Buyers, D.L. Abernathy, G.D. Samolyuk, G.M. Stocks and S.E. Nagler, "Quantum oscillations of nitrogen atoms in uranium nitride," Nature Communications 3, 1124 (2012).**

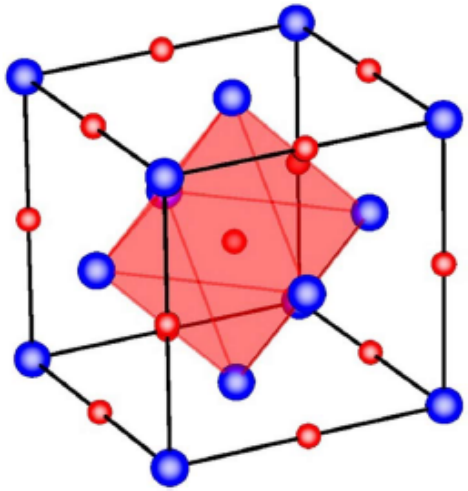

Figure 1 | Rocksalt crystal structure of uranium nitride. Each N atom (small red spheres) is centred in a regular octahedron of U atoms (large blue spheres).

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