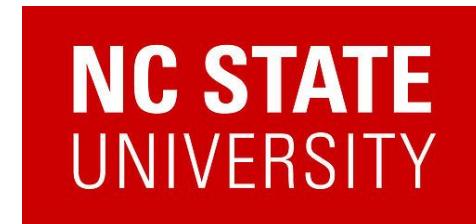


# Pendellösung Interferometry as a Probe for New Interactions

Benjamin Heacock  
NIST

PSI Workshop  
October 22, 2019



# Overview

Pendellosung Interferometry

Precise measurements of neutron-crystalline structure factors

Physics contained in crystalline structure factors

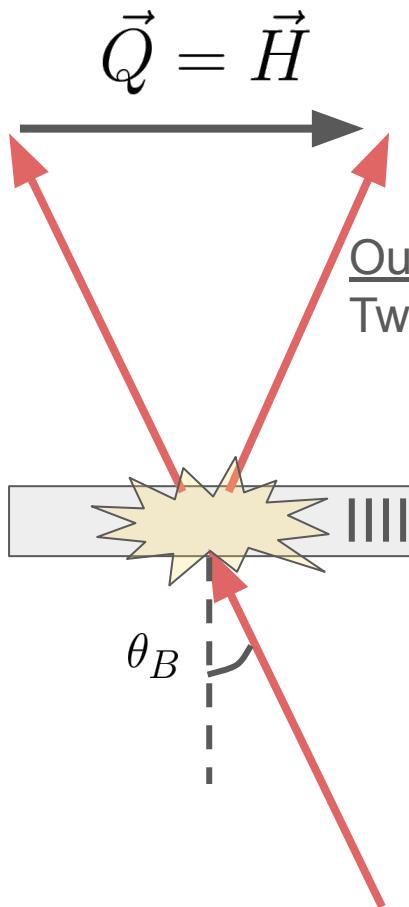
Thermal vibrations, neutron charge radius, BSM force mediators

What can precision measurements (relative unc  $\sim 5 \times 10^{-5}$ ) of neutron-silicon structure factors achieve?

Experiment

Preliminary Results

# Pendellösung - Bragg scattering as a two-state system



Momentum Transfer:  
Set by Bragg plane spacing

Outside the crystal:  
Two free neutron states

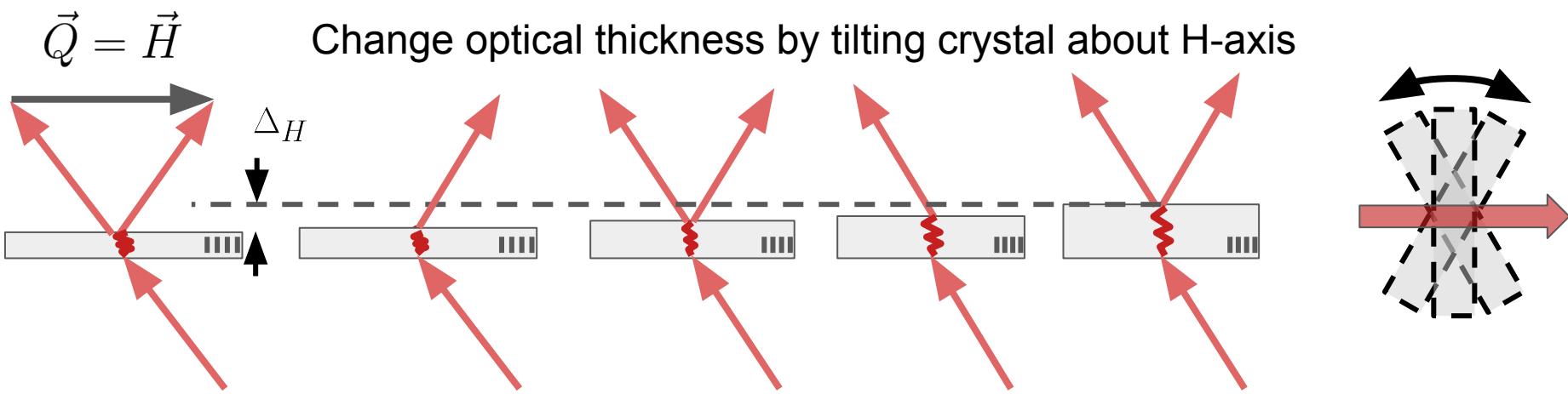
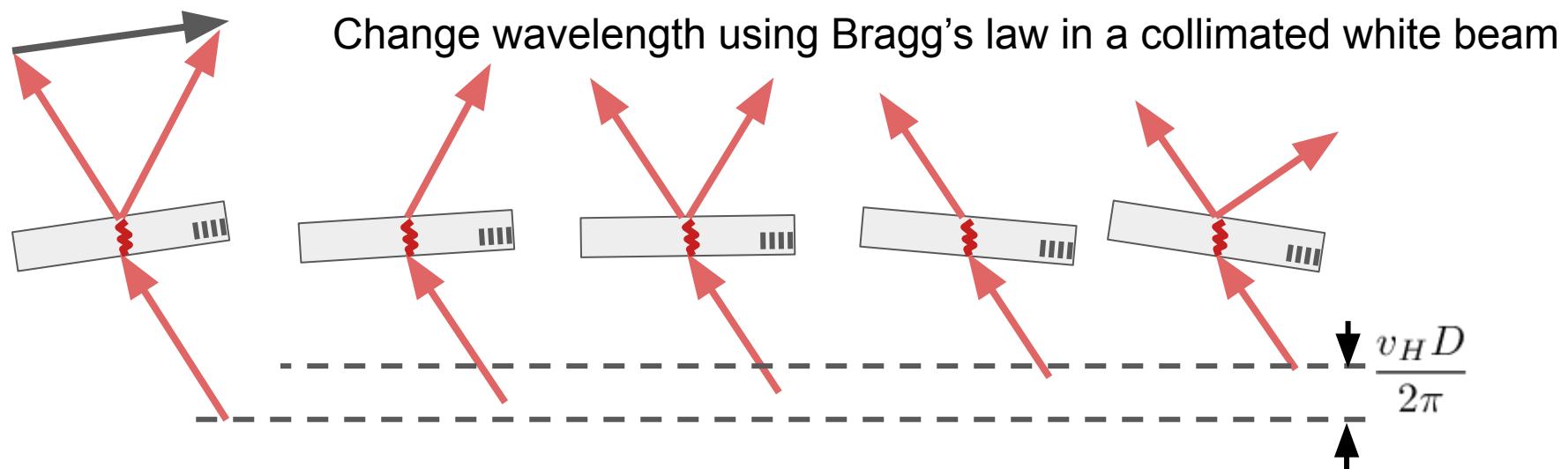
Inside the crystal:  
Two states  
Linear combinations of external states  
Each state sees separate index of refractions  
Same total energy, differing kinetic terms  
Pendellosung is the interference between two internal states

$$H = \frac{2\pi}{d} = \frac{2\pi}{a} \sqrt{h^2 + l^2 + k^2}$$

$$\mathcal{H} = \frac{\hbar^2}{2m} \begin{pmatrix} K^2 + v_0 & v_H \\ v_{-H} & K_H^2 + v_0 \end{pmatrix}$$

$$\vec{K}_H = \vec{K} + \vec{H}$$

# Measuring Pendelloosung



# Measuring Pendelosung

In Summary: By rotating a crystal in a carefully-prepared neutron beam, the phase of the oscillations between the diffracted and transmitted beams provides a precise measure of the neutron-crystalline potential.

# OBSERVATION OF PENDELLÖSUNG FRINGE STRUCTURE IN NEUTRON DIFFRACTION\*

C. G. Shull

Brookhaven National Laboratory, Upton, New York,  
and Massachusetts Institute of Technology, Cambridge, Massachusetts  
(Received 26 August 1968)

## Spherical-Wave Neutron Propagation and Pendellösung Fringe Structure in Silicon\*

C. G. Shull and J. A. Oberteuffer

Massachusetts Institute of Technology, Cambridge, Massachusetts 02139  
(Received 11 August 1972)

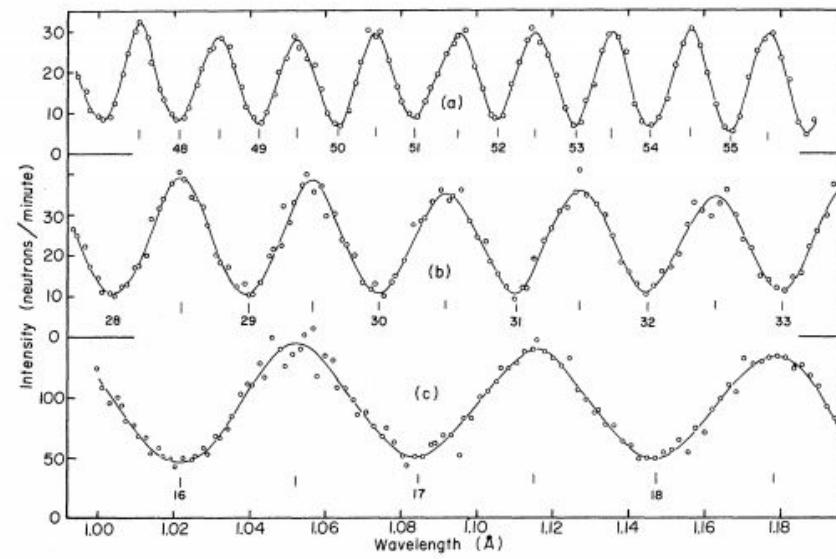
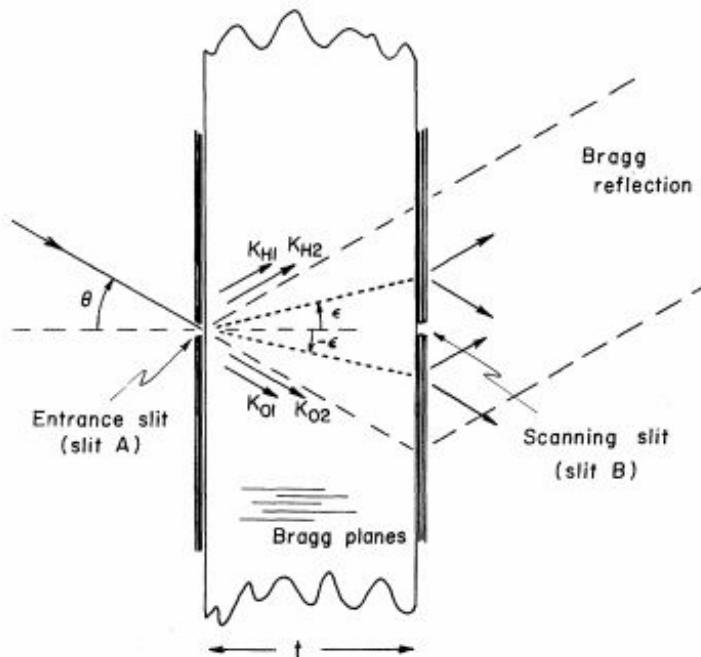
## Neutron Pendellösung Fringe Structure in the Laue Diffraction by Germanium \*

C. G. Shull and W. M. Shaw

Massachusetts Institute of Technology, Cambridge, Massachusetts, USA

(Z. Naturforsch. **28 a**, 657—661 [1973]; received 22 January 1973)

Dedicated to Gerhard Borrmann on his 65th birthday



# Pendellosung Phase Shift

Crystal structure factors

Fourier transform of the crystalline potential evaluated at  $Q=H$

Sensitive to physics on the atomic scale, including new physics → **BSM Force Mediator**

Probes atomic charge densities → **Neutron Charge Radius**, Schwinger scattering, nEDM

Crystalline Symmetry - Phase shift for each scattering center

Interference between scattering centers causes selection rules (forbidden reflections) and highly suppressed Schwinger scattering and nEDM terms for centrosymmetric crystals

Averaging the phases over thermal vibrations → **Debye Waller Factor**

# Pendellosung Phase Shift

VV Fedorov, Poster 40: Neutron spin rotation at Laue diffraction in a weakly deformed transparent noncentrosymmetric crystal

Crystal structure factors

Fourier transform of the crystalline potential evaluated at  $Q=H$

Sensitive to physics on the atomic scale, including new physics → BSM Force Mediator

Probes atomic charge densities → Neutron Charge Radius, Schwinger scattering, nEDM

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Averaging the phases over thermal vibrations → Debye Waller Factor

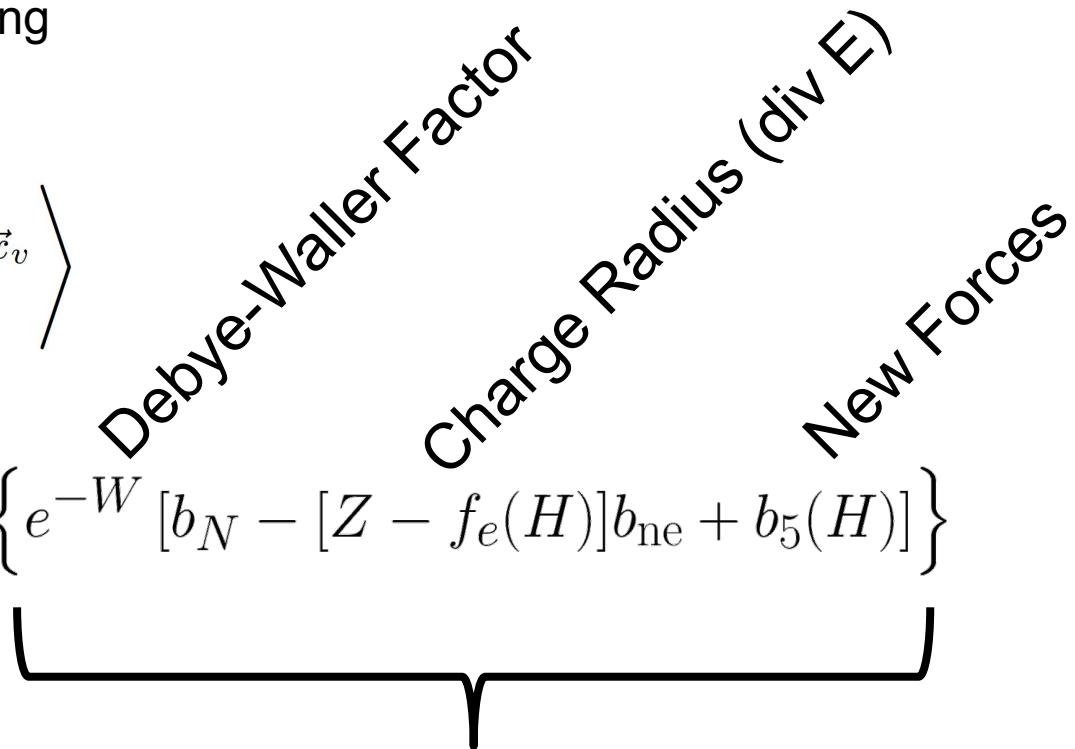
# Crystal Structure Factor

Neutron-atom  
interaction

Thermal  
smearing

$$v_H = \frac{2m}{a^3} \tilde{V}(H) \left\langle \sum_v e^{i\vec{H} \cdot \vec{x}_v} \right\rangle$$

$$v_H = \left( \frac{4\pi}{a^3} \sum_v e^{i\vec{H} \cdot \vec{x}_v} \right) \left\{ e^{-W} [b_N - [Z - f_e(H)]b_{ne} + b_5(H)] \right\}$$



# Debye-Waller Factor

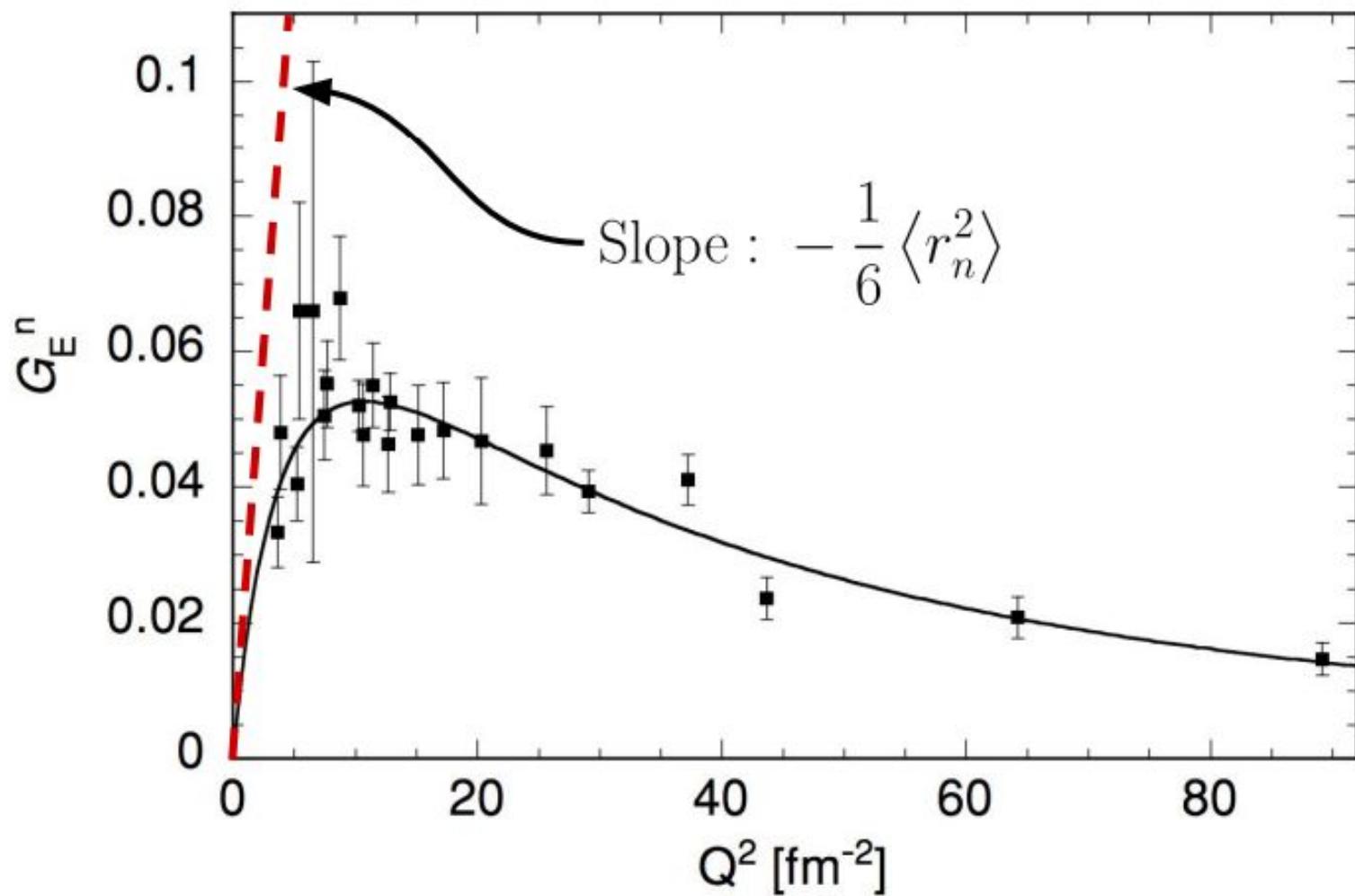
Mean-square displacement matrix takes on a simple form for centrosymmetric crystals

$$\left\langle e^{i\vec{H} \cdot \vec{u}} \right\rangle = e^{-\frac{1}{2}\vec{H} \cdot U \cdot \vec{H}}$$

$$U_{ij} = \frac{1}{3} \langle u^2 \rangle \delta_{i,j}$$

$$\left\langle e^{i\vec{H} \cdot \vec{u}} \right\rangle = e^{-W} = \exp \left[ -\frac{1}{16\pi^2} \textcircled{B} H^2 \right]$$

# Neutron Charge Radius



# Neutron charge radius deduced from Bragg reflection technique

J.-M. Sparenberg\* and H. Leeb

*Atominsttitut der Österreichischen Universitäten, Technische Universität Wien, Wiedner Hauptstraße 8-10, A-1040 Vienna, Austria*

(Received 16 January 2002; published 27 November 2002)

The possibility of the determination of the neutron mean square charge radius from high-precision thermal-neutron measurements of the nuclear scattering length and of the scattering amplitudes of Bragg reflections is considered. Making use of the same Pendellösung technique as Shull [Phys. Rev. Lett. **21**, 1585 (1968)], the scattering amplitudes of about eight higher-order Bragg reflections in silicon could be measured without single-scattering contamination. This would provide a value of the neutron charge radius as precise as the disagreeing Argonne-Garching and Dubna values, as well as a Debye-Waller factor of silicon ten times more precise than presently available.

DOI: 10.1103/PhysRevC.66.055210

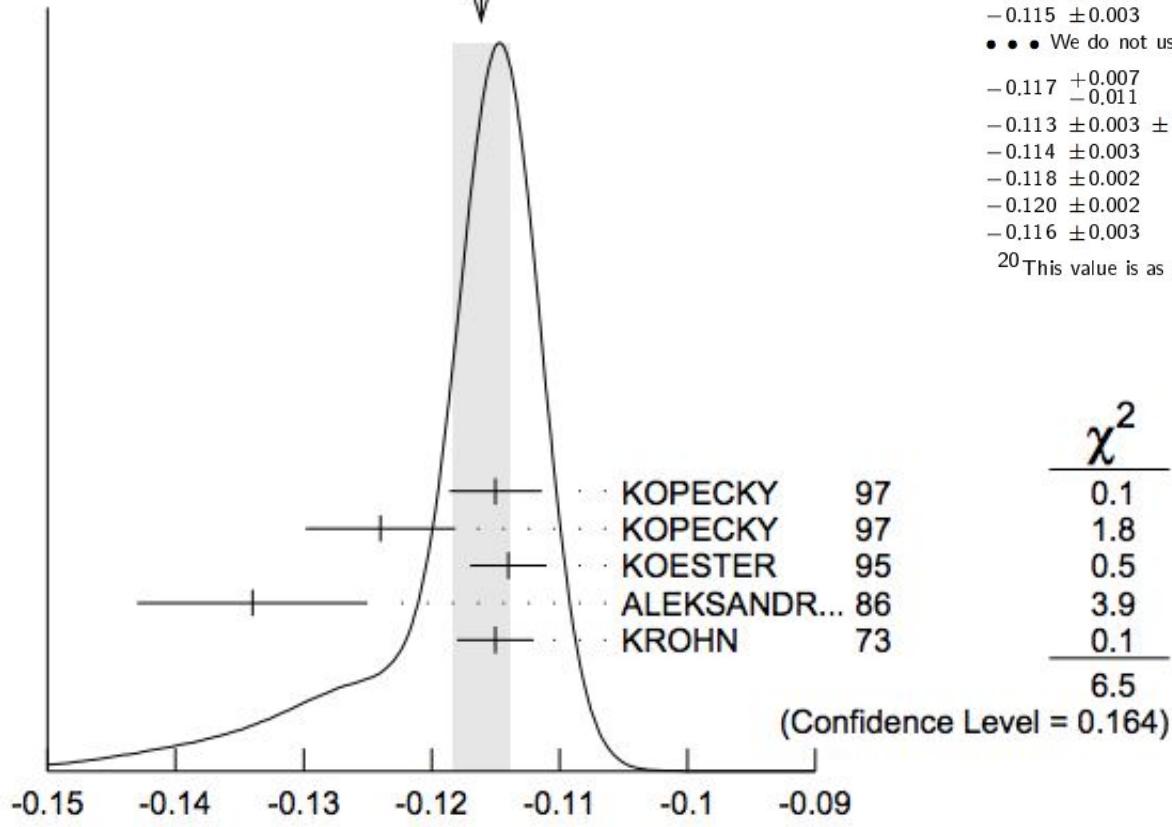
PACS number(s): 14.20.Dh, 03.75.Dg, 13.60.Fz, 28.20.Cz

hkl	$f_{\text{x-ray}}$	$b(Q)/b(0)-1$
111	10.6298(5)	-0.01094(20)
220	8.4179(6)	
311	7.7134(10)	
400	7.0293(4)	
331	6.7571(9)	
•	•	
•	•	
•	•	
880	1.5514(18)	

## $n$ MEAN-SQUARE CHARGE RADIUS

# PDG Values

WEIGHTED AVERAGE  
 $-0.1161 \pm 0.0022$  (Error scaled by 1.3)



The mean-square charge radius of the neutron,  $\langle r_n^2 \rangle$ , is related to the neutron-electron scattering length  $b_{ne}$  by  $\langle r_n^2 \rangle = 3(m_e a_0 / m_n) b_{ne}$ , where  $m_e$  and  $m_n$  are the masses of the electron and neutron, and  $a_0$  is the Bohr radius. Numerically,  $\langle r_n^2 \rangle = 86.34 b_{ne}$ , if we use  $a_0$  for a nucleus with infinite mass.

VALUE (fm <sup>2</sup> )	DOCUMENT ID	COMMENT
<b><math>-0.1161 \pm 0.0022</math> OUR AVERAGE</b>		Error includes scale factor of 1.3. See the ideogram below.
$-0.115 \pm 0.002 \pm 0.003$	KOPECKY 97	$ne$ scattering (Pb)
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$-0.116 \pm 0.003$	KROHN 66	$ne$ scattering (Ne, Ar, Kr, Xe)

<sup>20</sup>This value is as corrected by KOESTER 76.

x-ray form factor

$$[Z - f_e(Q)] b_{ne}$$

$$b_{ne} = \frac{\langle r_n^2 \rangle}{86.34 \text{ fm}}$$

# Neutron interferometric method to provide improved constraints on non-Newtonian gravity at the nanometer scale

Geoffrey L. Greene

*Department of Physics, University of Tennessee, Knoxville, Tennessee 37996, USA and Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*

Vladimir Gudkov

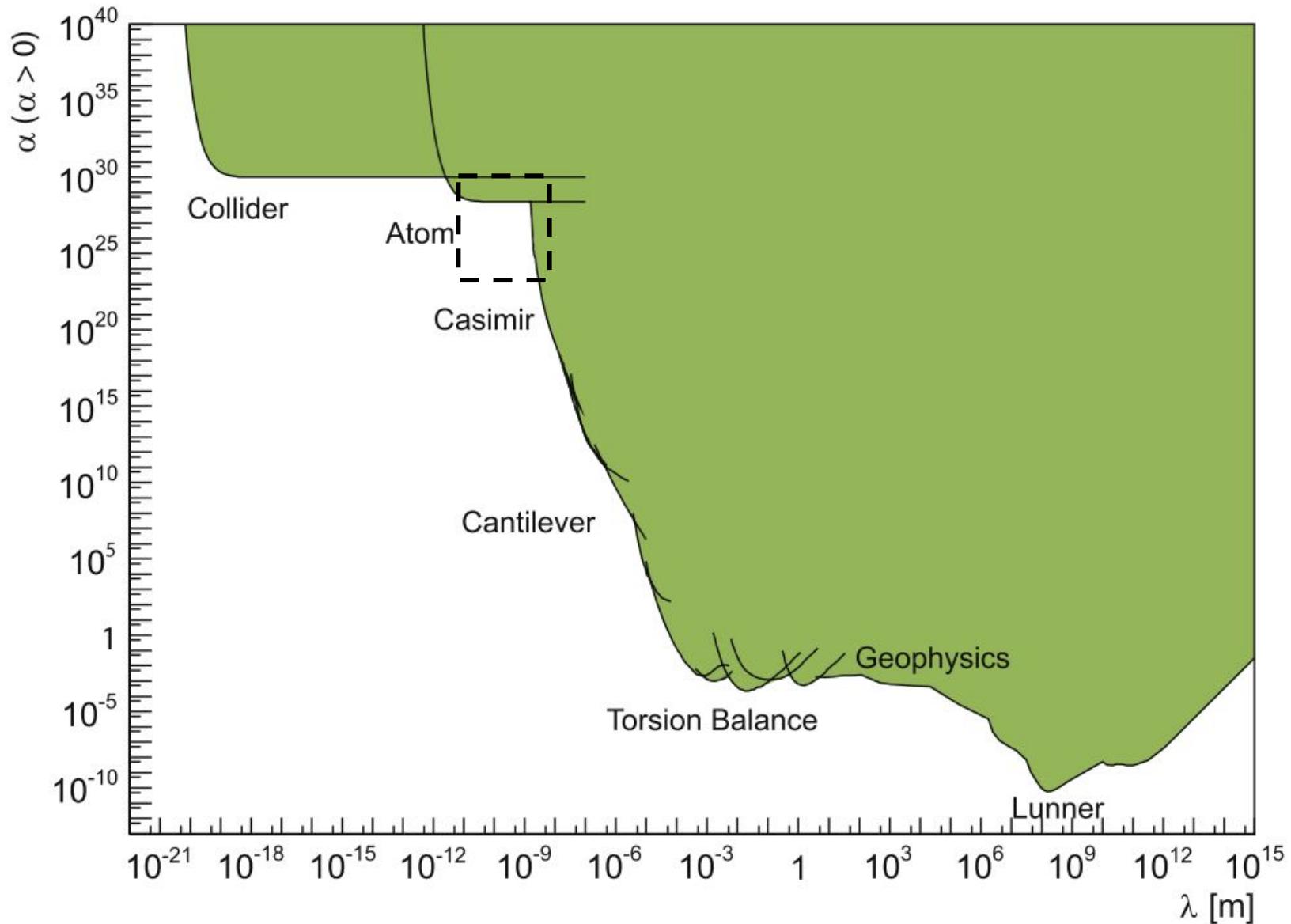
*Department of Physics and Astronomy, University of South Carolina, Columbia, South Carolina 29208, USA*

(Received 31 August 2006; published 4 January 2007)

In recent years, an energetic experimental program has set quite stringent limits on a possible “non- $1/r^2$ ” dependence on gravity at short length scales. This effort has been largely driven by the predictions of theories based on compactification of extra spatial dimensions. It is characteristic of many such theories that the strength and length scales of such anomalous gravity are not clearly determined from first principles. As a result, it is productive to extend the current limits the range and strength of such hypothetical interactions. As a heavy, neutral, and (almost) stable particle, the neutron provides an ideal probe for the study of such hypothetical interactions at very short range. In this work, we describe methods based on neutron interferometry which have the capability to provide improved sensitivity non-Newtonian forces down to length scales at and below an nanometer.

# Theoretical Motivations for Short-Range Forces

- General Relativity (GR) is non-renormalizable
  - It breaks down at high energies
  - Weakest of four forces and very high energies → Difficult to measure
  - Gravitational phenomena has never been measured at a length scale less than  $10 \mu\text{m}$
- Some theories that provide an ultraviolet (UV) completion to GR also predict short range forces
  - Perturbations to gravity at less than  $100 \mu\text{m}$
  - Many theories → wide parameter space to constrain
- Other new physics predict new short range forces
  - Light Dark Matter (Fayet 2007)
  - Dark Energy (Zee 2004)
  - Neutrino Masses (Arkani-Hamed & Dimopoulos 2002)



# New Forces

## Yukawa (Most Common)

$$V = -G \frac{m_1 m_2}{r} \left[ 1 + \alpha e^{-r/\lambda} \right]$$

$$b_5(Q) = -\frac{m}{2\pi} g^2 Q_n Q_N \frac{\lambda_5^2}{1 + Q^2 \lambda^2} = \alpha_G 2Gm^2 M \left( \frac{\lambda_5^2}{1 + Q^2 \lambda_5^2} \right)$$

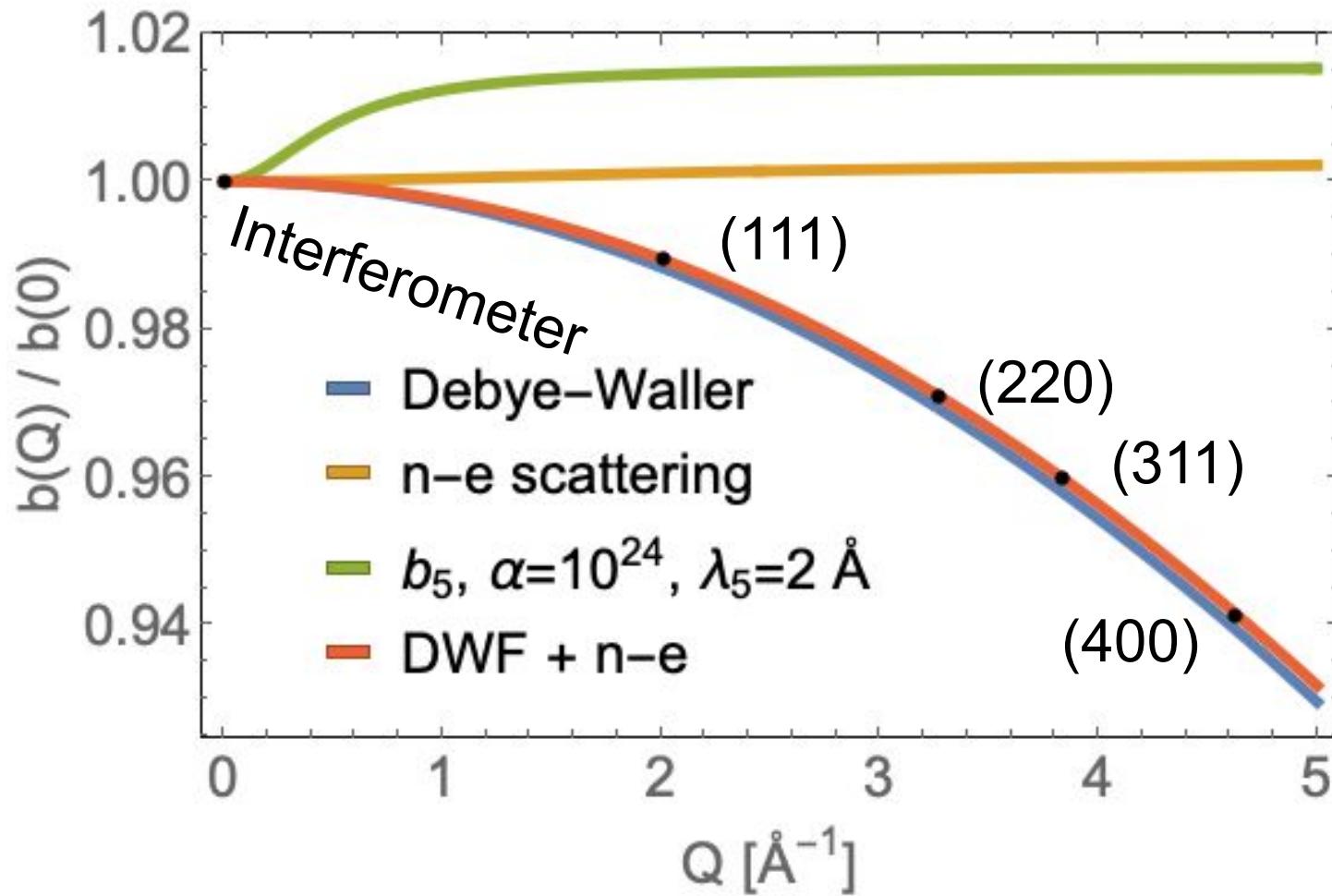
$$b_5(Q) = -1.6 \times 10^{-26} \alpha_G \frac{\lambda_5^2}{1 + Q^2 \lambda_5^2}$$

(Silicon)

Power Law Arkani-Hamed, Dimopoulos, and Dvali (ADD)

$$V = -G \frac{m_1 m_2}{r} \left[ 1 + \left( \frac{\lambda}{r} \right)^n \right]$$

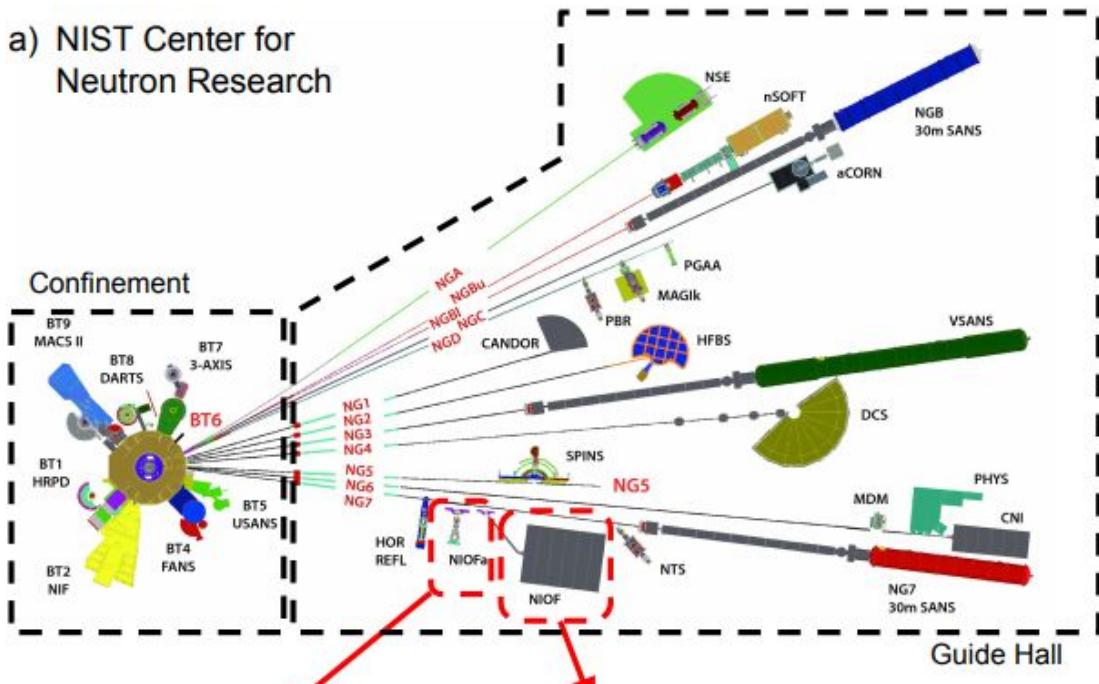
$$b(Q) = \exp \left[ -\frac{1}{16\pi^2} B Q^2 \right] \{ b_N - [Z - f_e(Q)] b_{\text{ne}} + b_5(Q) \}$$



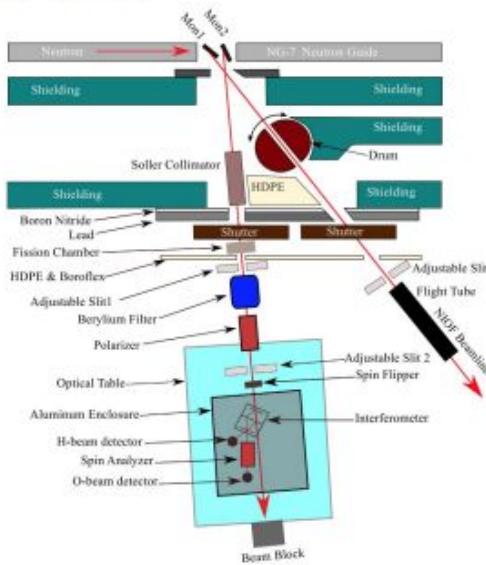
# NIST Center for Neutron Research



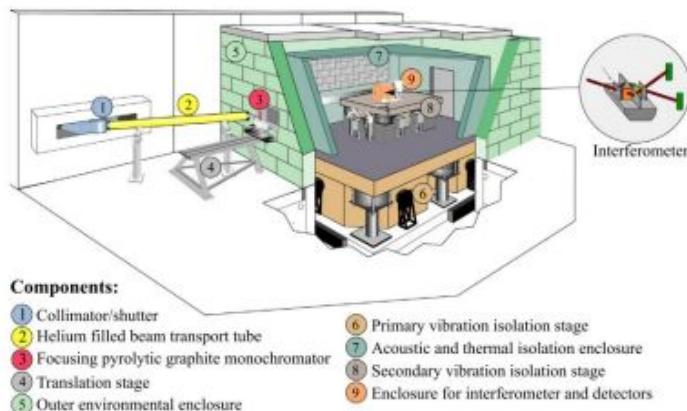
## a) NIST Center for Neutron Research



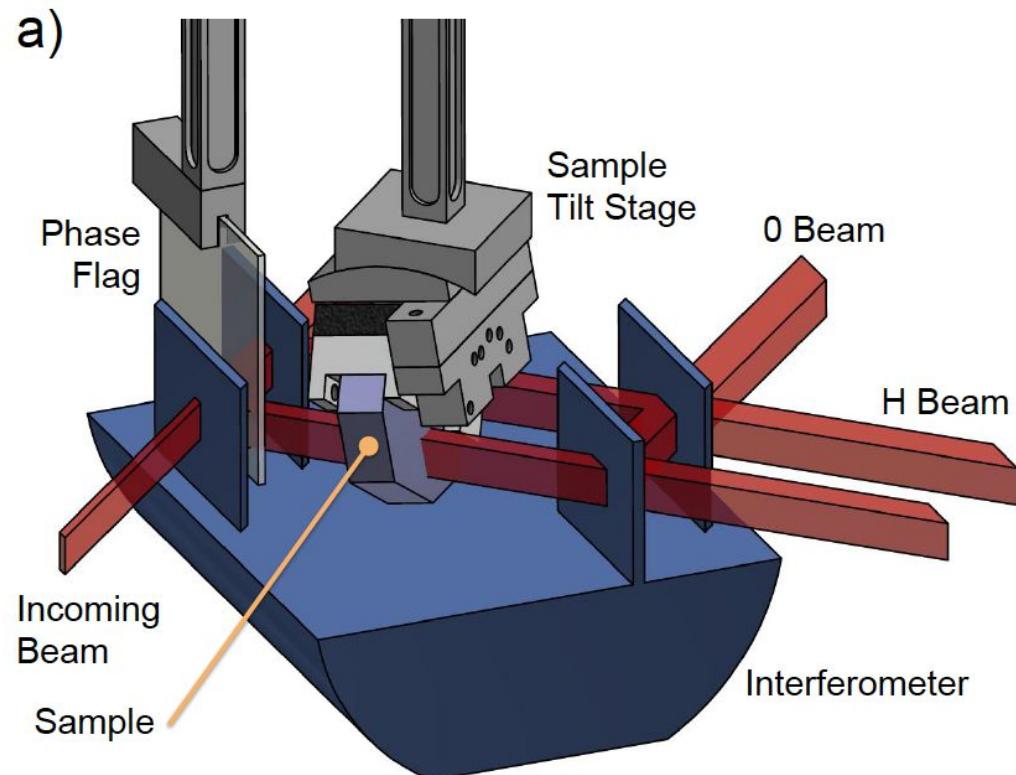
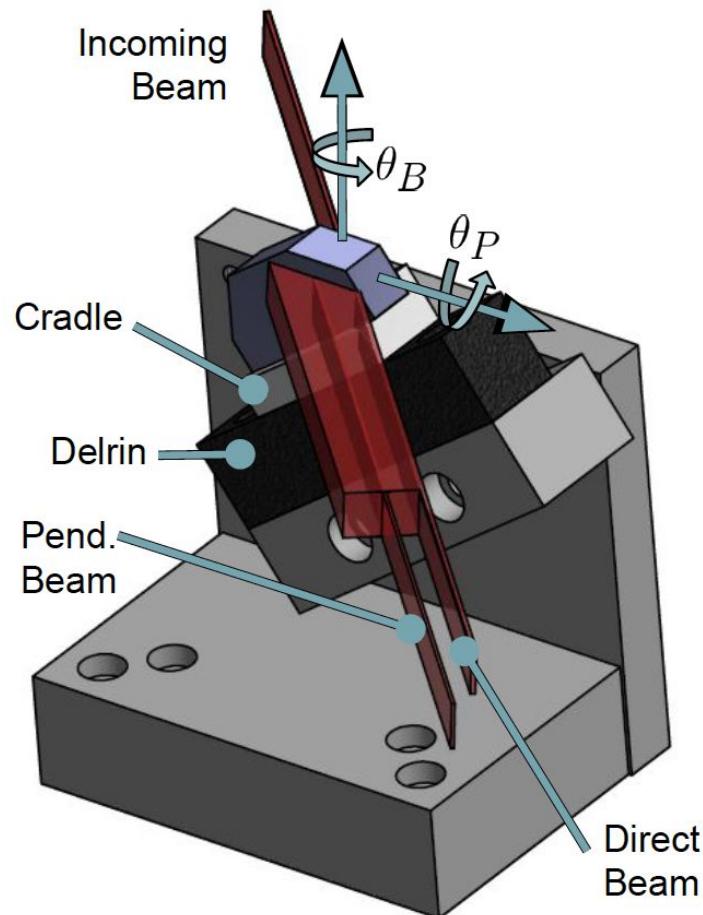
b) NIOFa



c) NIOF



# Experiment



Measure  $D b(H)$

Measure  $D b(0)$

# Strain and Crystal Thickness

Measured phase shift is proportional to crystal thickness

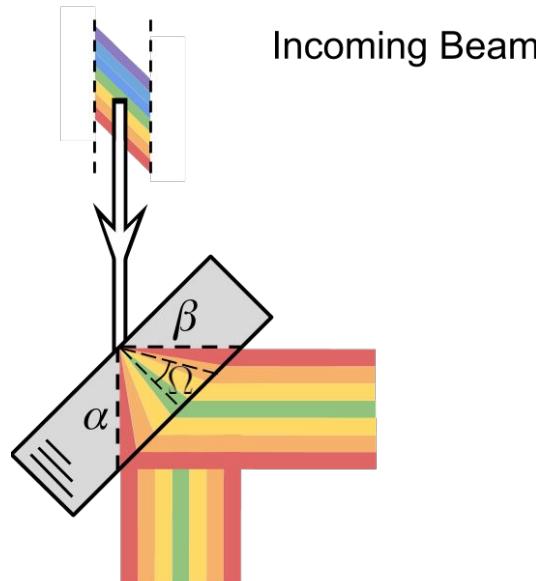
Strain gradients cause wavelength-dependent shift in the pendellosung fringe position

Previous measurements showed large strain gradients ( $\sim 100 \text{ nrad mm}^{-1}$ )

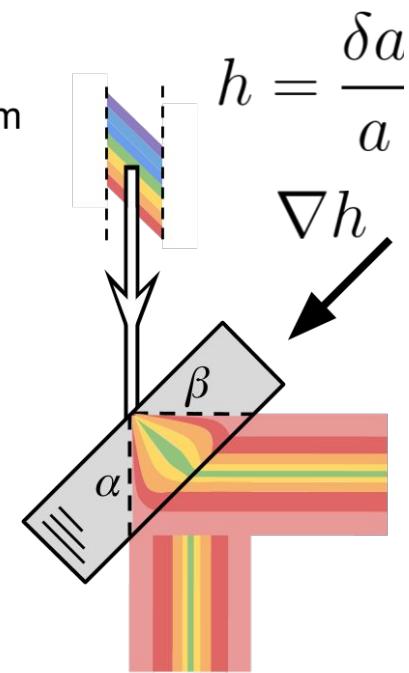
This level of strain is (relatively) large compared to experience with interferometers

**Required crystal flatness is at odds with need to chemically etch**

## Normal Trajectories



## Strained Trajectories



# Strain and Crystal Thickness

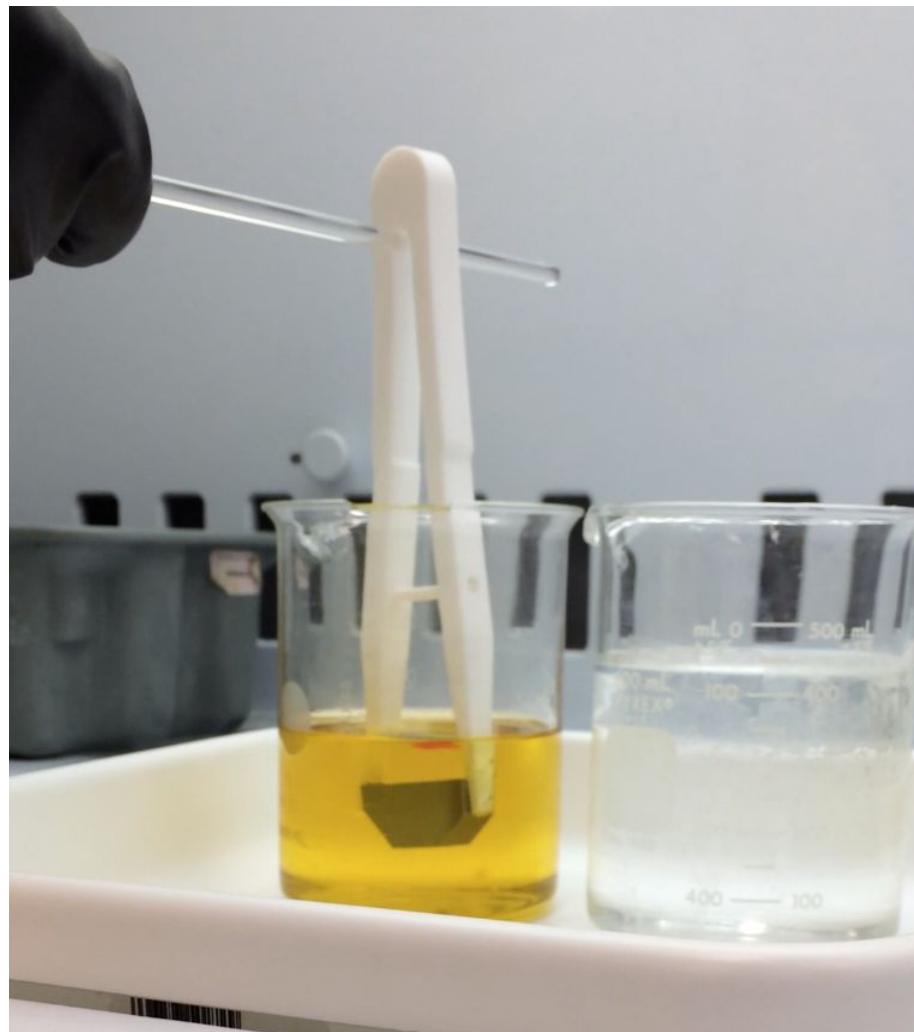
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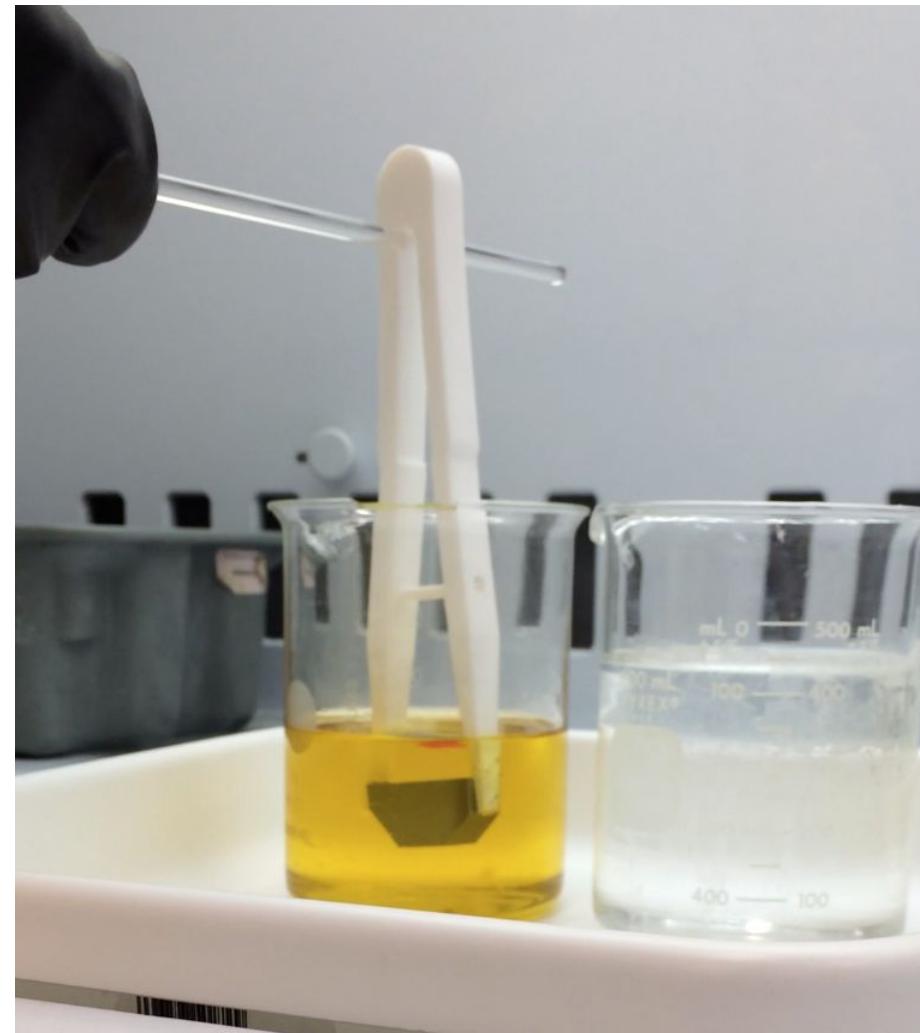
This level of strain is (relatively) large compared to experience with interferometers

**Required crystal flatness is at odds with need to chemically etch**



# Strain and Crystal Thickness

Interferometer	Condition	$\nabla h$ ( $10^{-9}$ mm $^{-1}$ )
Skew	Etched (>30 um)	4
"Thin Blade"	Etched (>30 um)	5
	Annealed	1
Two-Blade	Precision Machined	$>10^4$
	Annealed	8
	Etched (4 um)	0.2
Shull	"Polished"	100



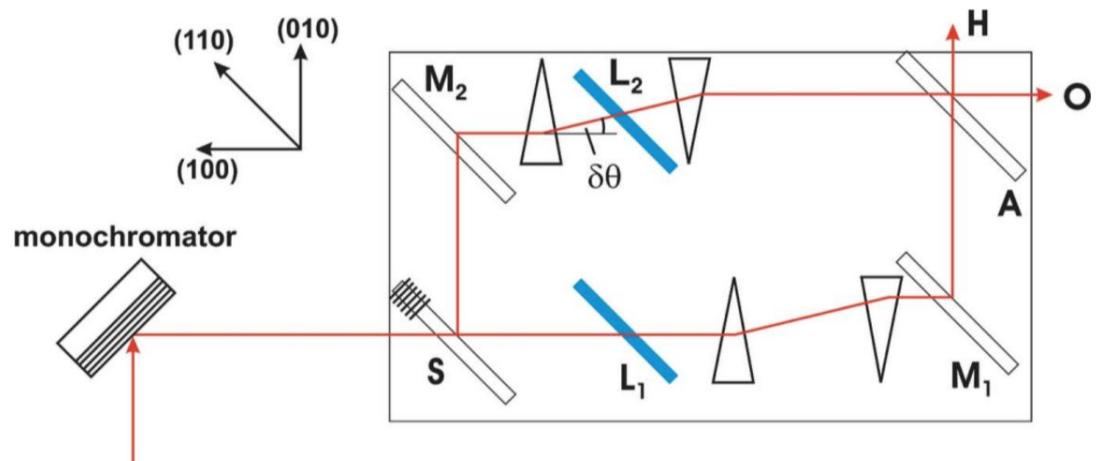
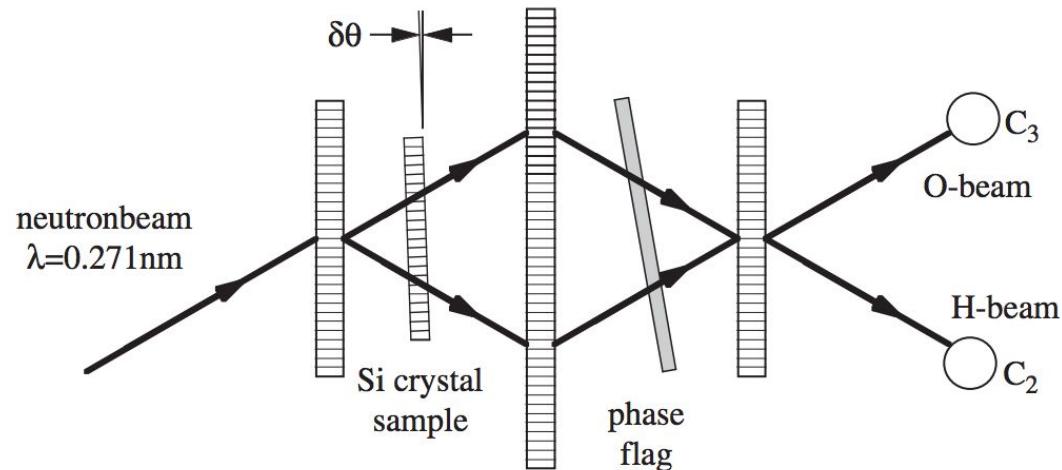
# Interferometer Inspiration

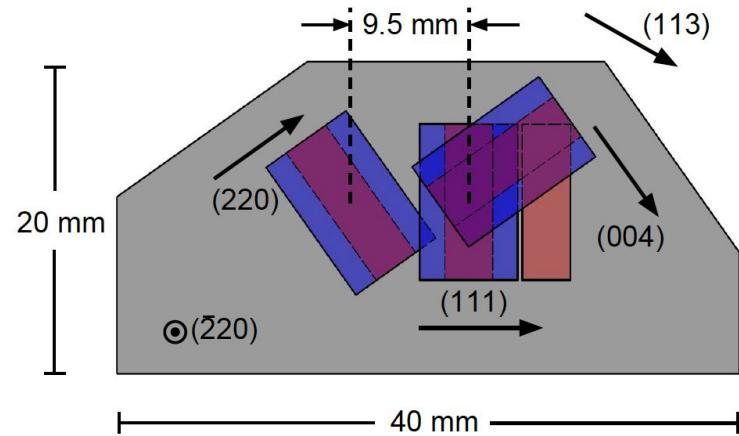
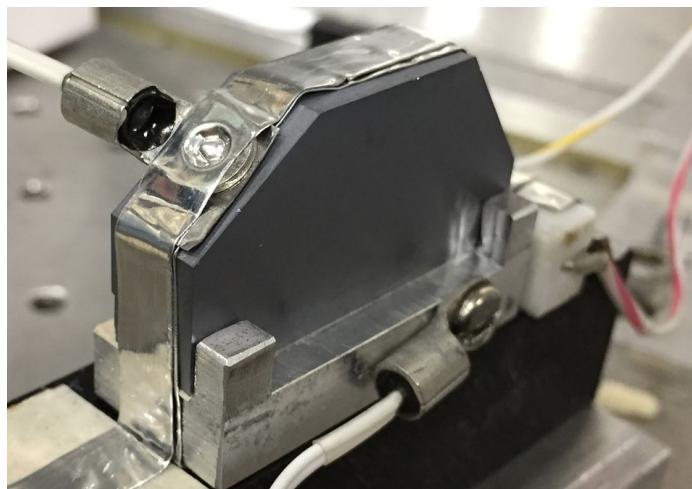
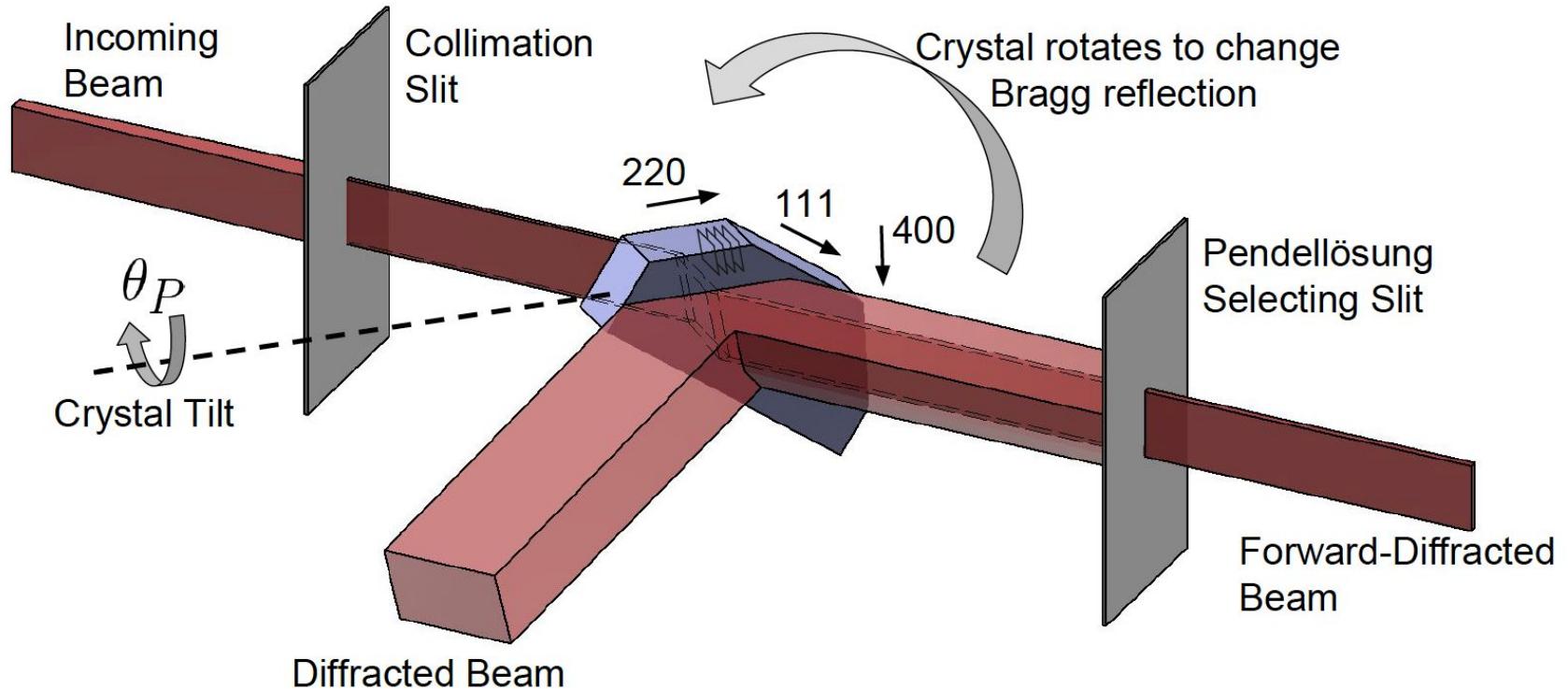
Sparenburg & Leeb 2003: -  
Pendellosung for multiple  $Q^2$

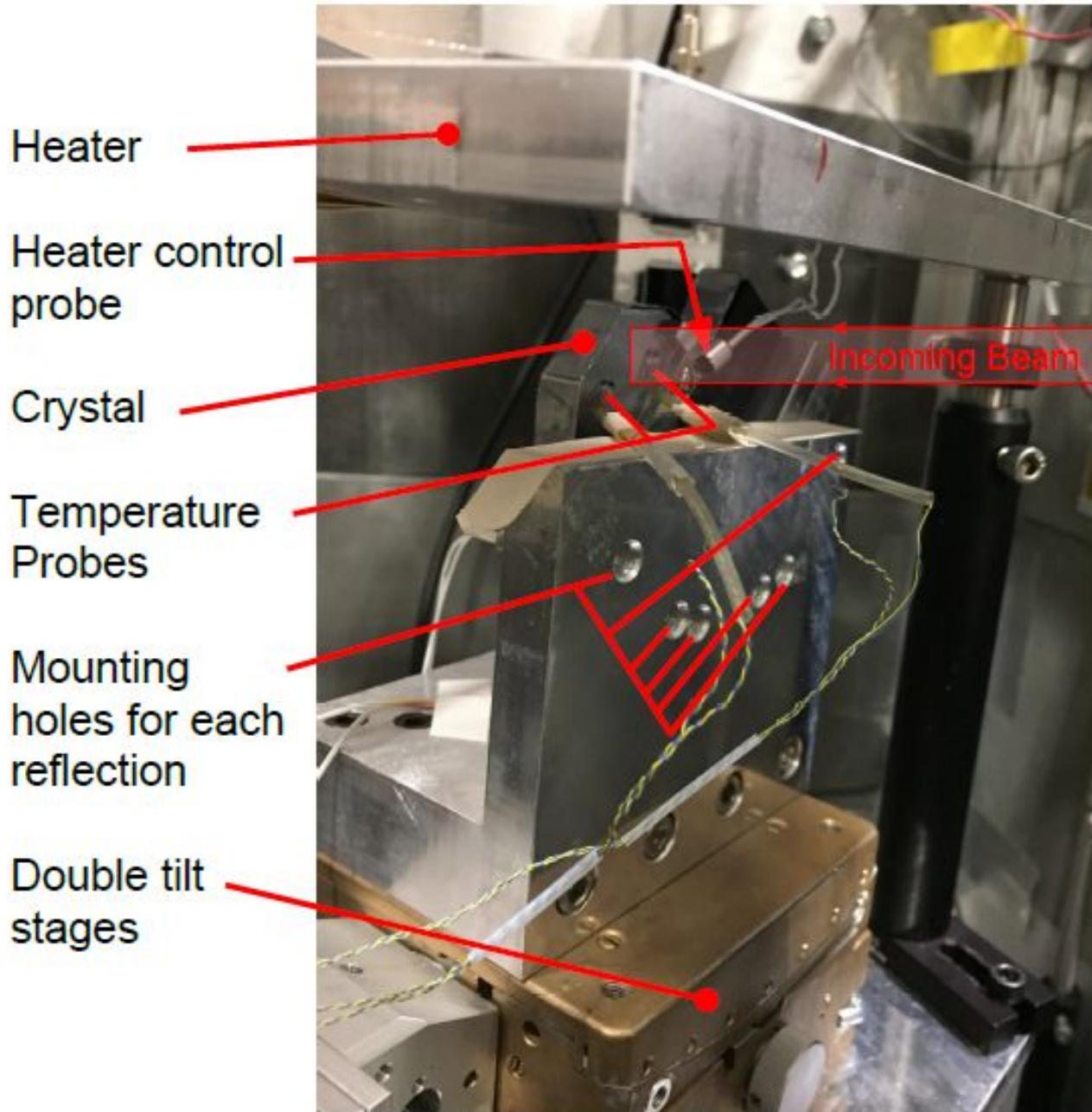
Wietfeldt et al. 2006\*: Diffracting  
crystal rotating in an  
interferometer

ILL Two Loop Interferometer\*

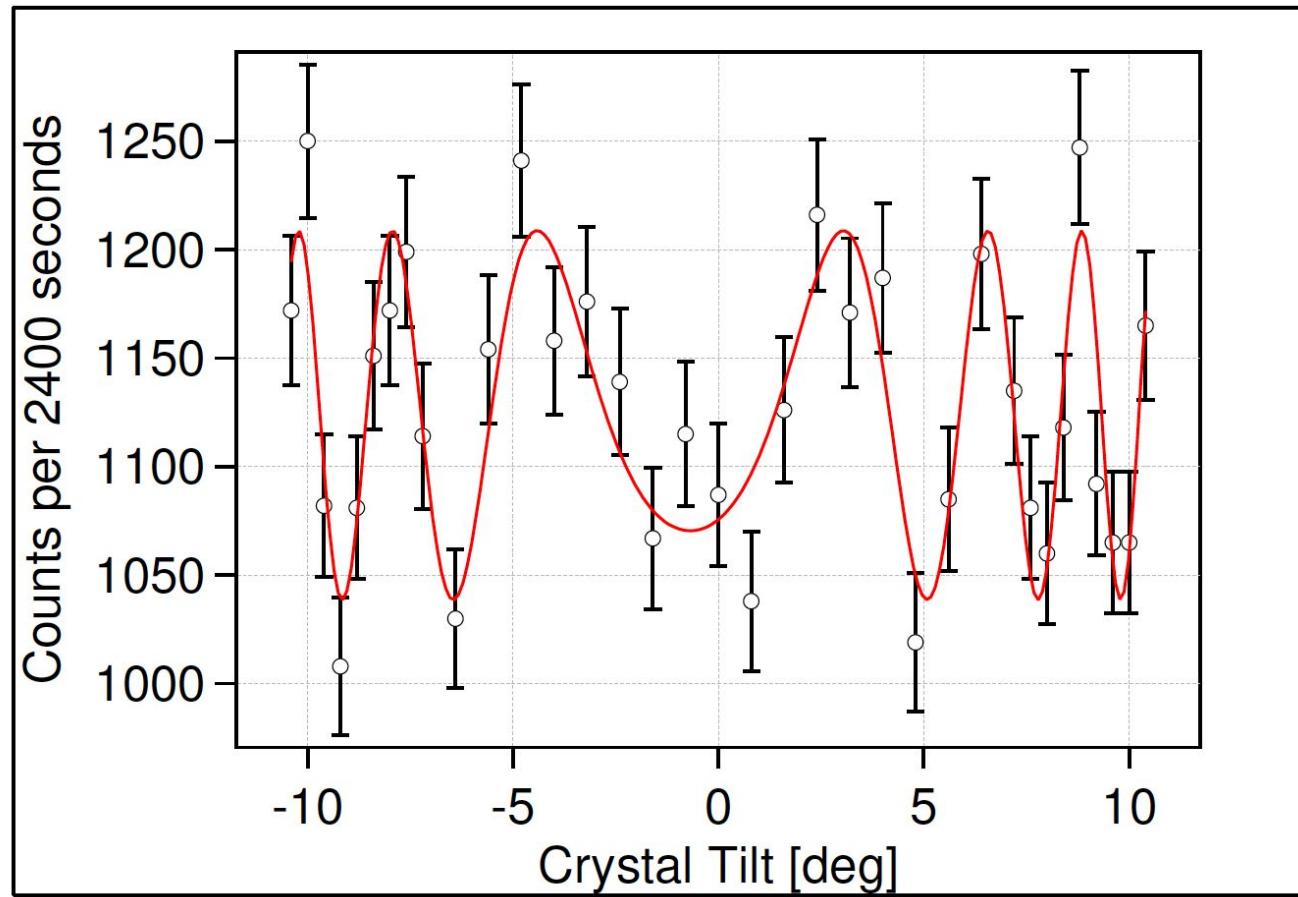
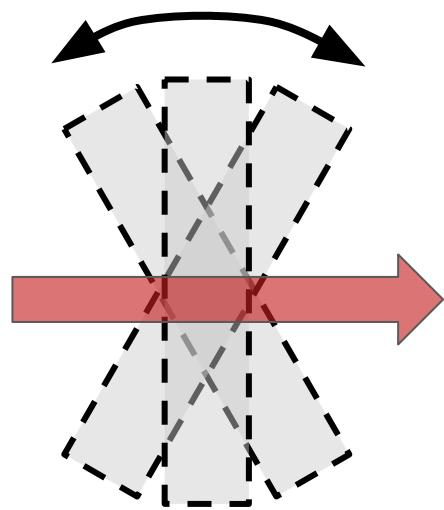
\*Relies on dynamical phase  
contributions from (twisted)  
interferometers

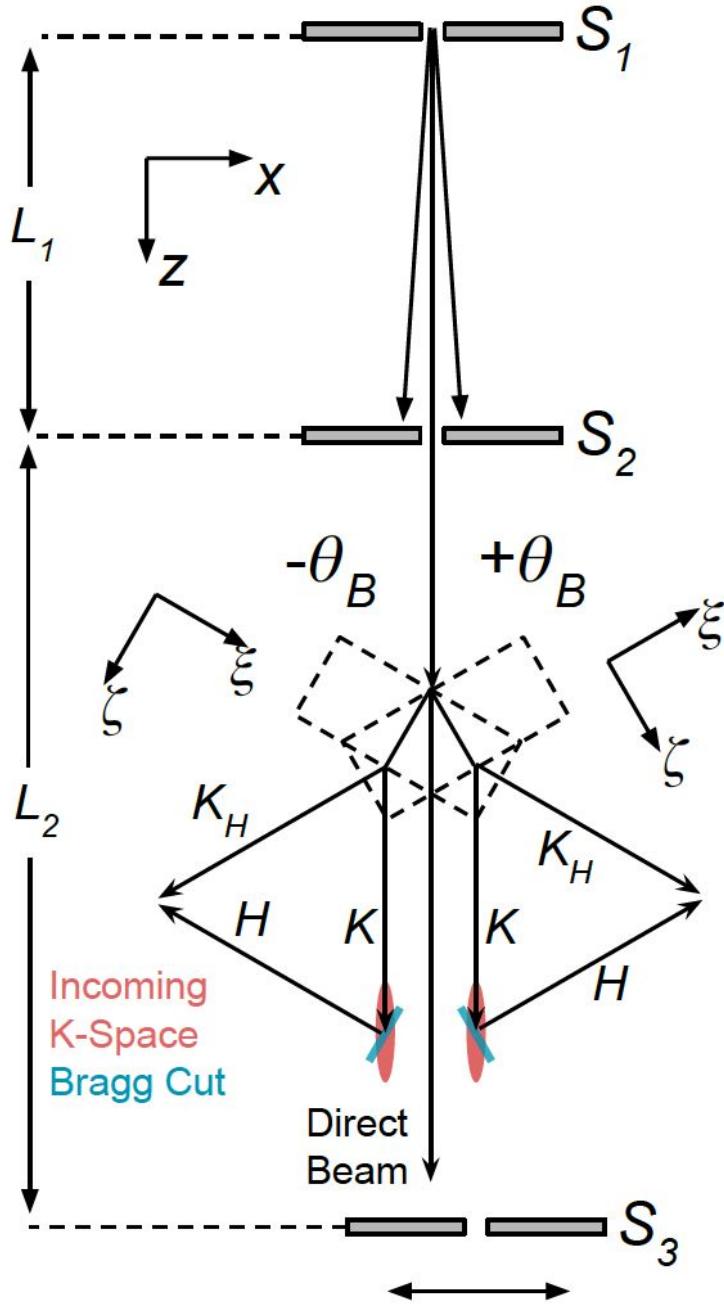






# Tilt to change optical thickness, constant $\lambda$





# Wavelength

Pendellosung phase shift proportional to  $\tan\theta_B$

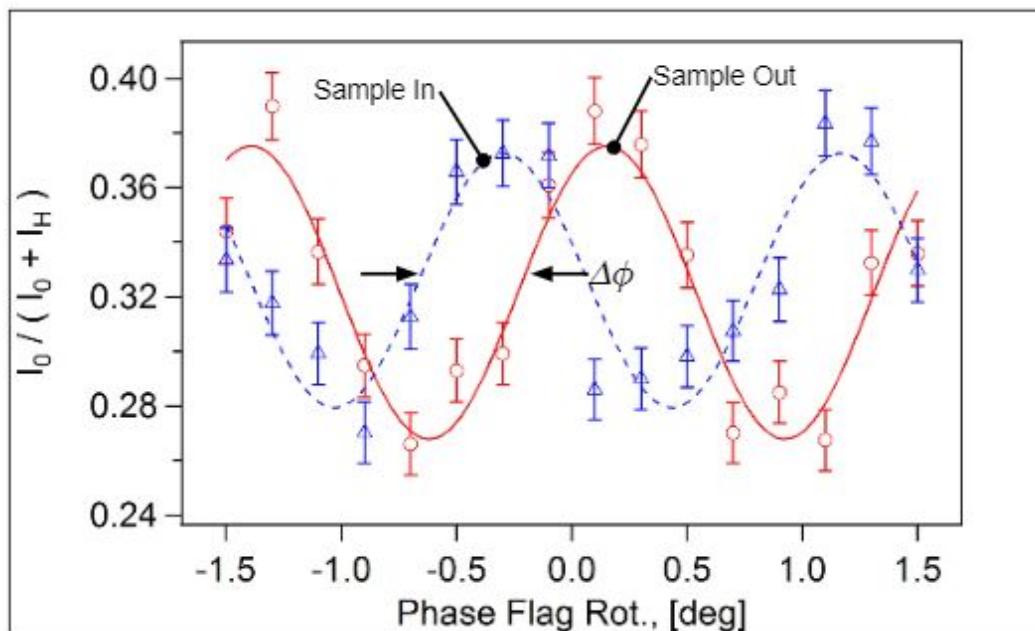
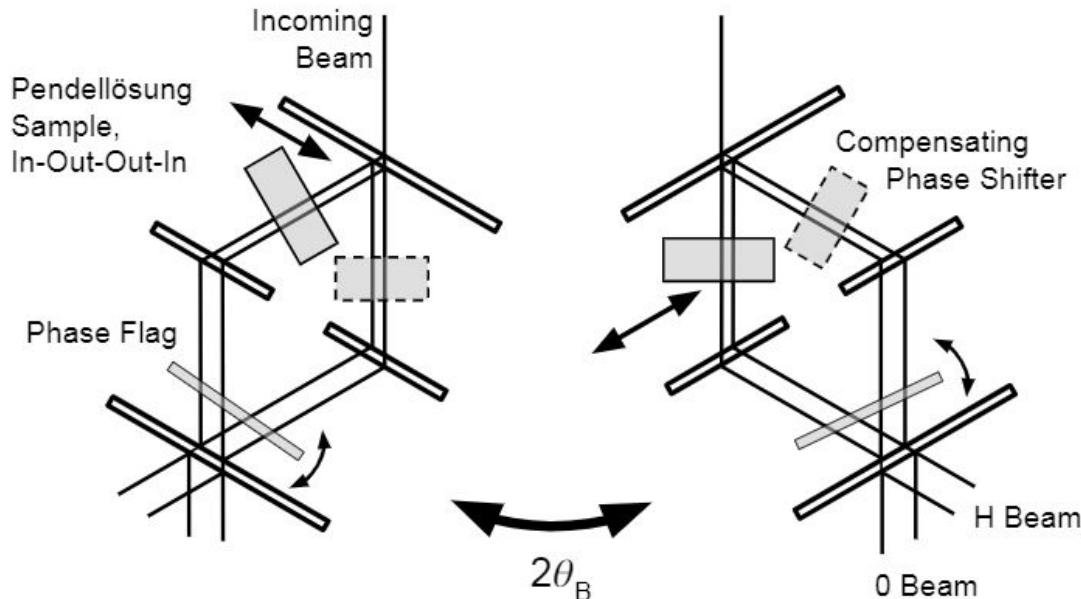
Measure at  $\pm\theta_B$

Average quadratic in  $\delta\theta_B$

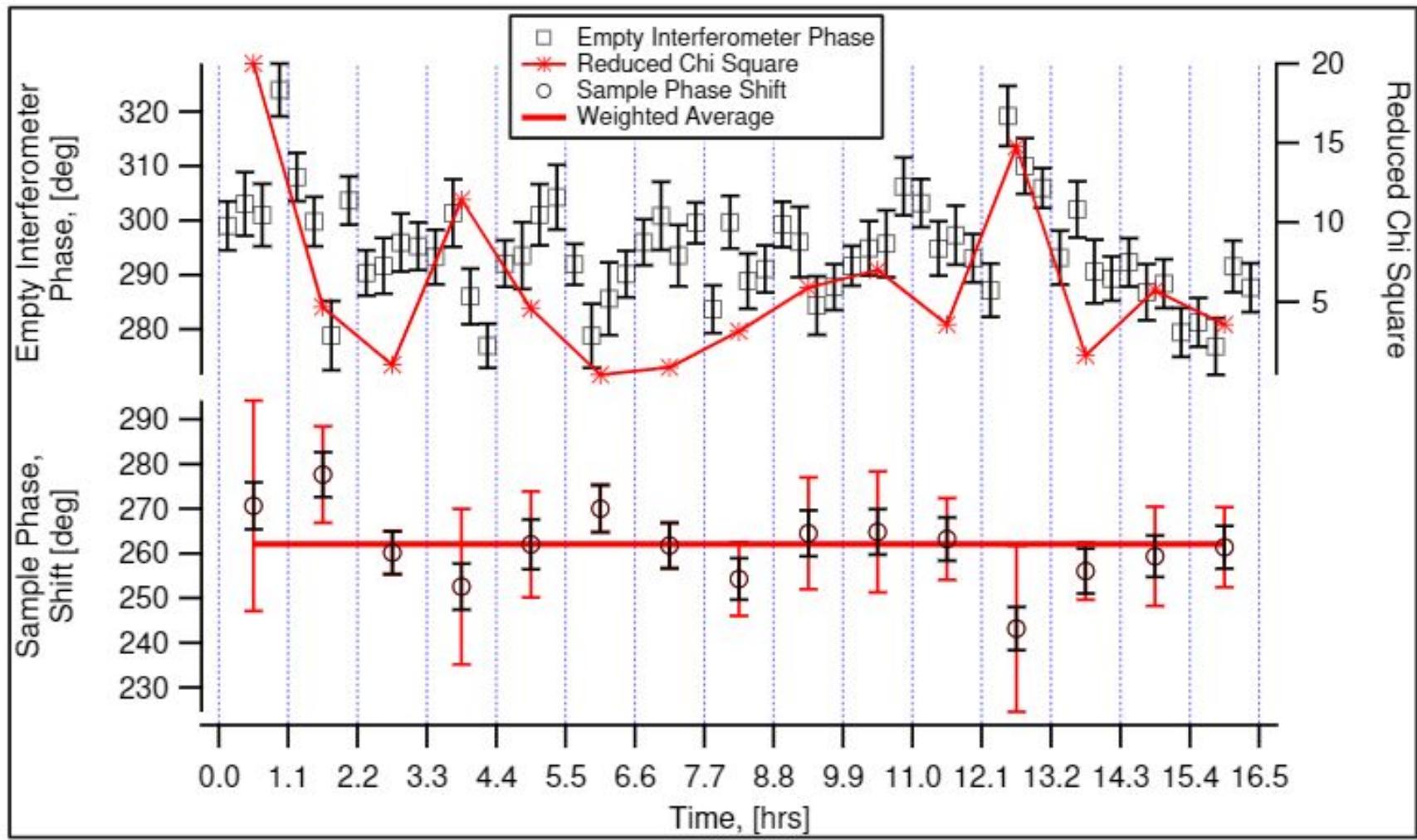
# Interferometer

In-Out-Out-In to subtract drifts

Repeated at  $\pm\theta_B$  to measure wavelength in-situ



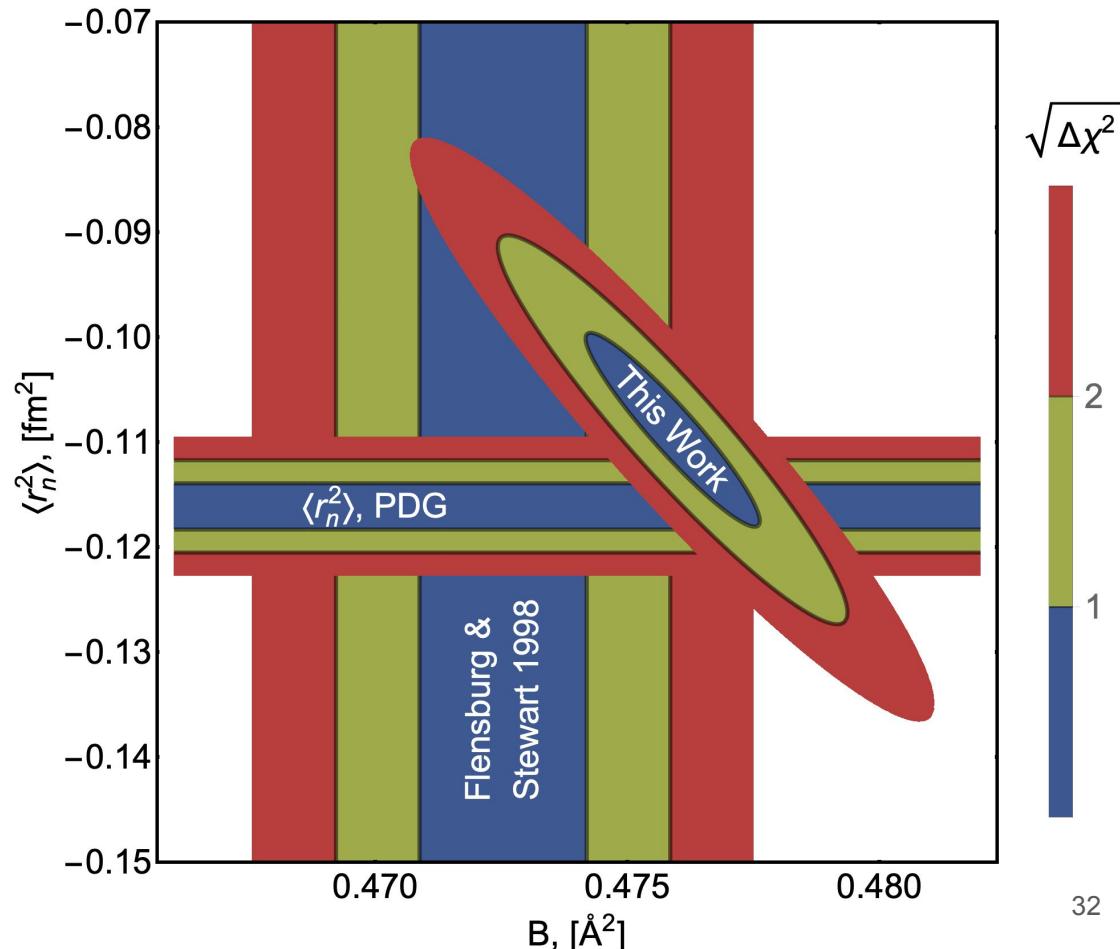
# Phase Shift and Drifts



# Preliminary Results

$hkl$	$f_{x\text{-ray}}$	$b(Q)/b(0)-1$
111	10.6298(5)	-0.01094(20) <b>-0.011084(55)</b>
220	8.4179(6)	<b>-0.030216(69)</b>
311	7.7134(10)	
400	7.0293(4)	<b>-0.060559(95)</b>
331	6.7571(9)	
422	6.1382(5)	
333	5.8041(6)	
•	•	
•	•	
•	•	
753	2.5508(29)	
•	•	
•	•	
880	1.5514(18)	

$hkl$	$b(Q)/b(0) - 1$	Unc. Stat.	Unc. Sys.	Unc. Total
(111)	-0.011 084	0.000 038	0.000 040	0.000 055
(220)	-0.030 216	0.000 059	0.000 036	0.000 069
(400)	-0.060 559	0.000 080	0.000 051	0.000 095

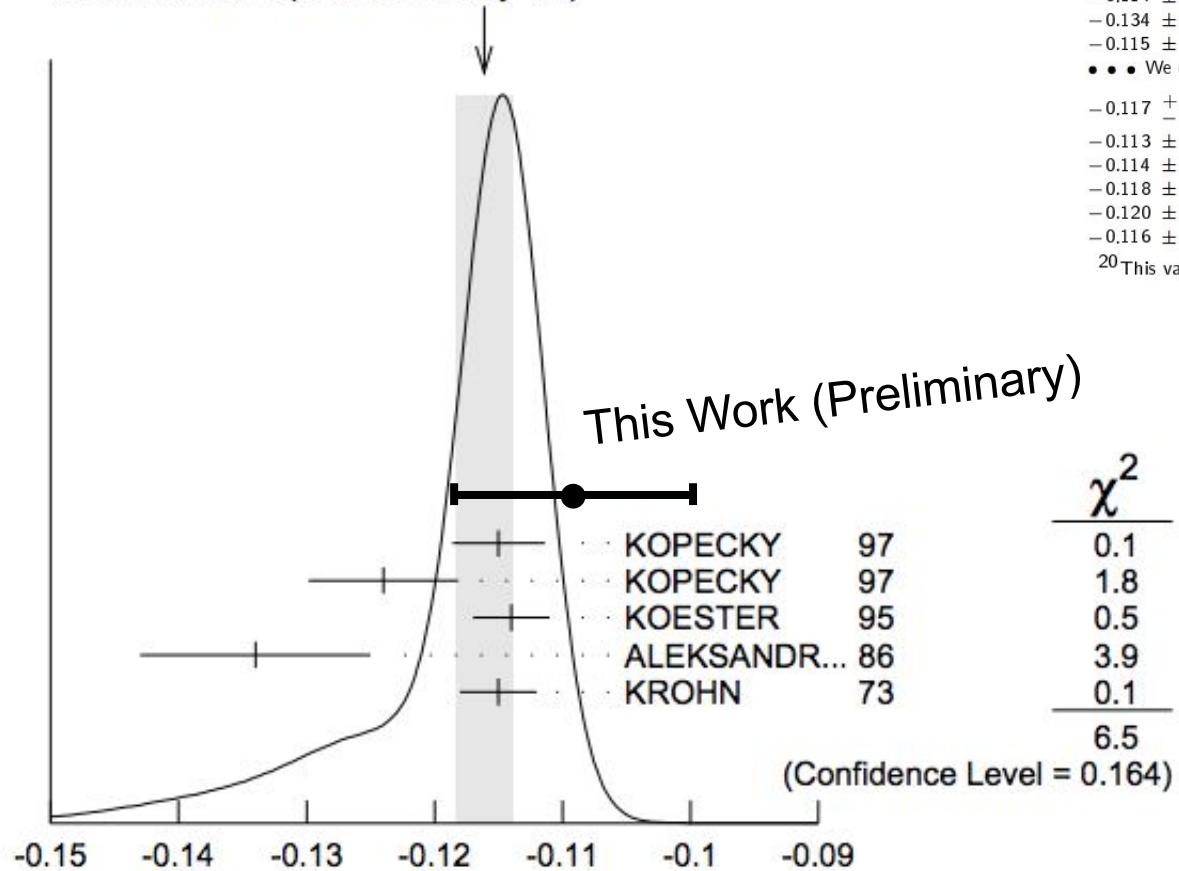


# Neutron Charge Radius

## $\langle r_n^2 \rangle$ MEAN-SQUARE CHARGE RADIUS

The mean-square charge radius of the neutron,  $\langle r_n^2 \rangle$ , is related to the neutron-electron scattering length  $b_{ne}$  by  $\langle r_n^2 \rangle = 3(m_e a_0 / m_n) b_{ne}$ , where  $m_e$  and  $m_n$  are the masses of the electron and neutron, and  $a_0$  is the Bohr radius. Numerically,  $\langle r_n^2 \rangle = 86.34 b_{ne}$ , if we use  $a_0$  for a nucleus with infinite mass.

WEIGHTED AVERAGE  
 $-0.1161 \pm 0.0022$  (Error scaled by 1.3)

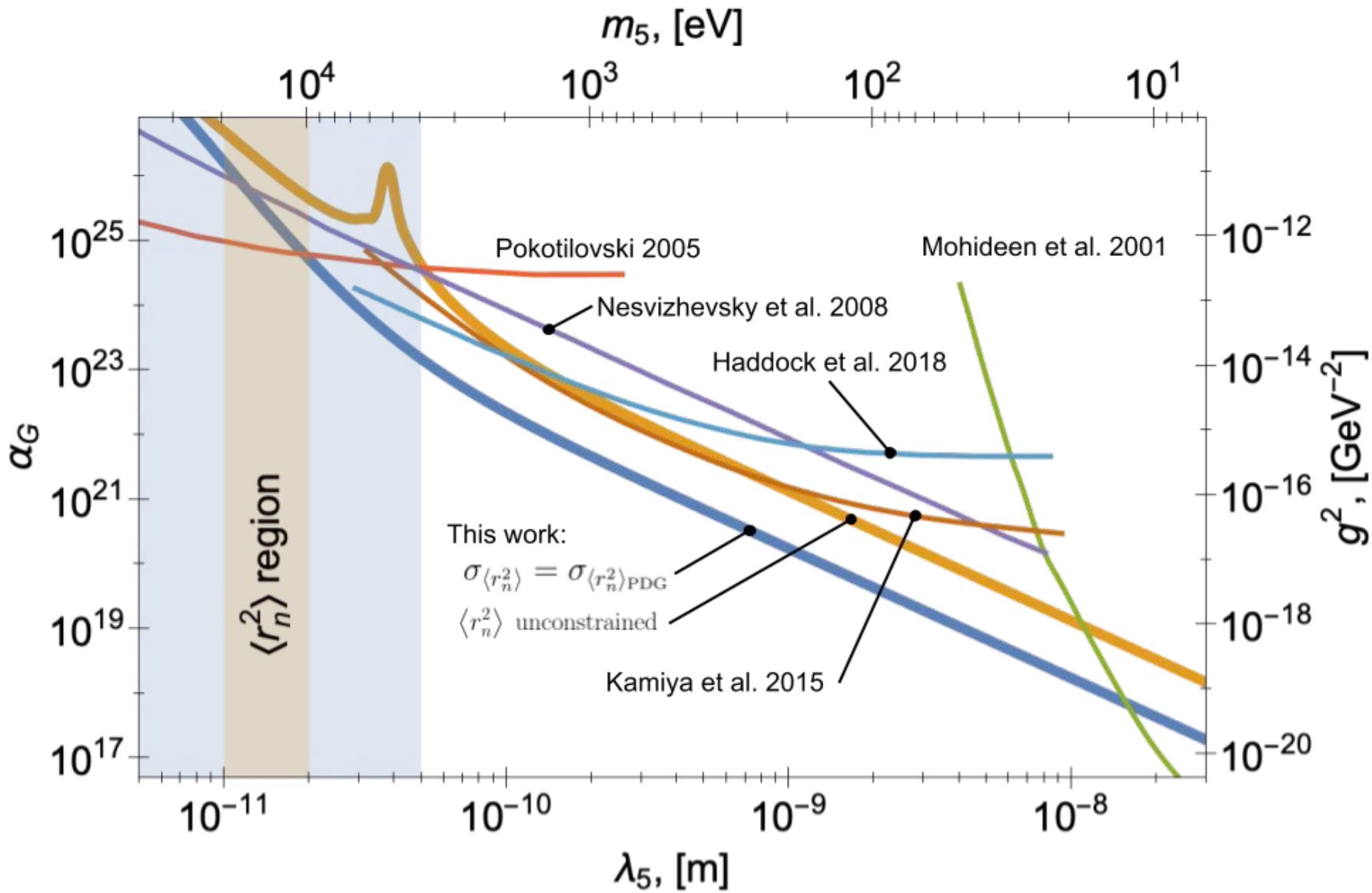


$n$  mean-square charge radius

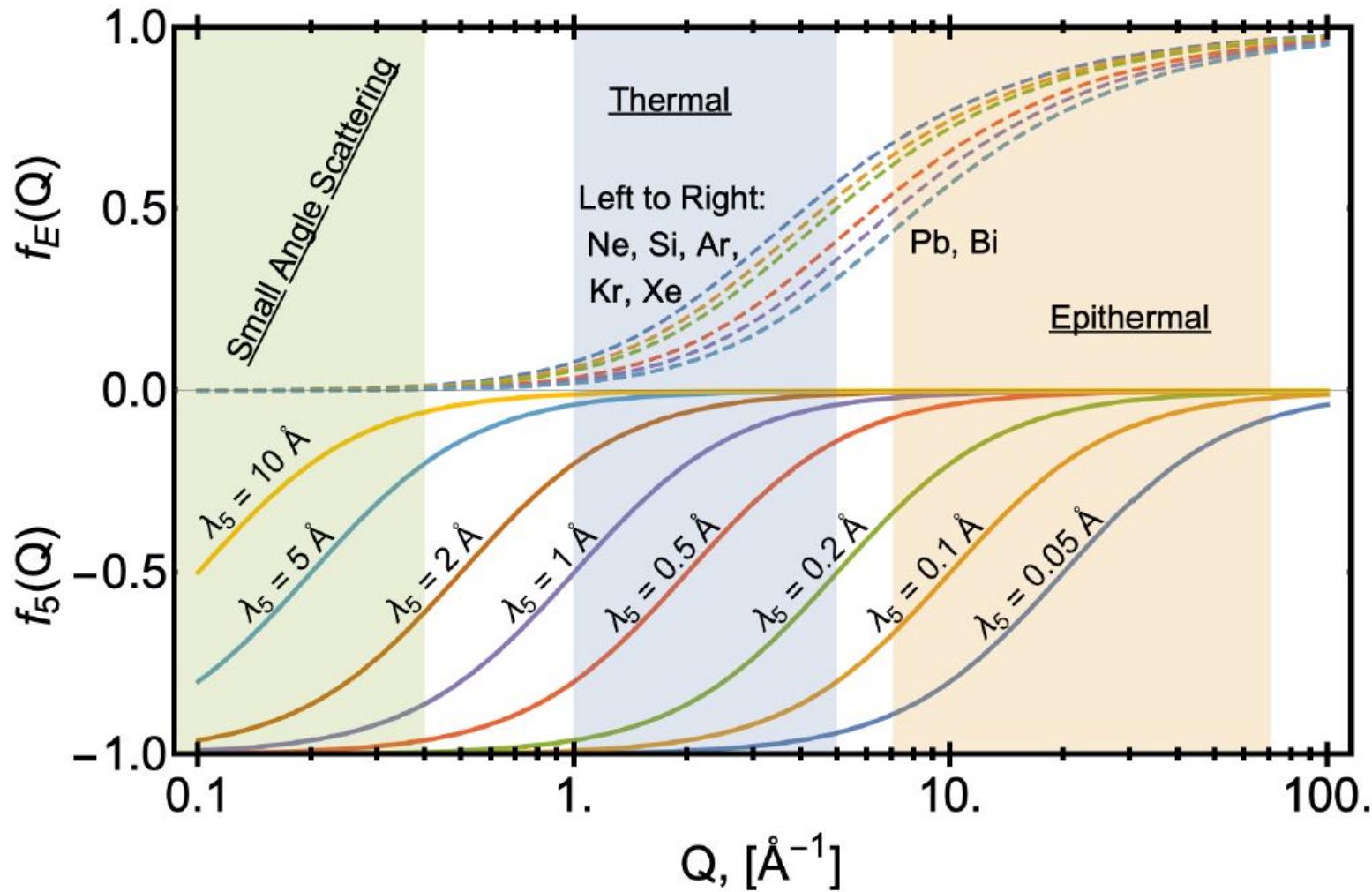
VALUE (fm <sup>2</sup> )	DOCUMENT ID	COMMENT
<b><math>-0.1161 \pm 0.0022</math> OUR AVERAGE</b>		Error includes scale factor of 1.3. See the ideogram below.
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-0.115 $\pm 0.003$	20 KROHN 73	$ne$ scattering (Ne, Ar, Kr, Xe)
• • • We do not use the following data for averages, fits, limits, etc. • • •		
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-0.120 $\pm 0.002$	KOESTER 76	$ne$ scattering (Bi)
-0.116 $\pm 0.003$	KROHN 66	$ne$ scattering (Ne, Ar, Kr, Xe)

<sup>20</sup> This value is as corrected by KOESTER 76.

# Preliminary Limits



# Length Scales and Atomic Form Factors



# Uncertainty Budget Pendelosung

Source	Uncertainty	$\Delta\phi_P$ (deg)	$\sigma_P/\phi_P \times 10^{-5}$	$hkl$
$S_3$ Translation	10 $\mu\text{m}$	0.0	0.5	111
		1.1	0.4	220
		2.9	0.3	400
Bragg Stage, $\theta_B$	$3 \times 10^{-4}$ deg		1.0	111
			0.8	220
			1.6	400
Temperature Gradient, $p_T^2/6$	$0.1 \times 10^{-5}$	0.2	0.1	111
	$0.2 \times 10^{-5}$	0.4	0.2	220
	$2.1 \times 10^{-5}$	3.7	2.1	400
Absolute Temperature	0.20 K	-1.0	0.7	111
	0.06 K	-1.0	0.6	220
	0.08 K	3.2	1.5	400
Crystal Profile	0.14 $\mu\text{m}$	1.4	1.4	111
Anharmonic Correction, $f_{\text{anh}}(hkl)$	$1 \times 10^{-5}$	2.9	1.0	111
Total Sys.			2.2	111
			1.1	220
			3.1	400
$\phi_P$ , Stat.	2.6 deg		2.5	111
	3.3 deg		5.4	220
	6.3 deg		7.8	400
Total			<b>3.0</b>	111
			<b>5.5</b>	220
			<b>8.4</b>	400

# Uncertainty Budget Interferometer

Source	Uncertainty	$\Delta\phi_I$ (deg)	$\sigma_I/\phi_I \times 10^5$	$hkl$
Crystal Translation	1 mm		2.4	111
			2.6	220
			3.8	400
Bragg Stage, $\theta_B$	$3 \times 10^{-4}$ deg		0.7	All
Air Scattering, $\phi_{air}$	0.8 deg	105.3 deg	1.6	All
Thermal phase, $\phi_{therm}$	0.9 deg	0.6 deg	1.7	All
<b>Total Sys.</b>			3.5	111
			3.5	220
			4.5	400
$\phi_I$ , Stat.	1.5 deg		2.9	111
	1.5 deg		2.9	220
	1.8 deg		3.5	400
<b>Total</b>			<b>4.4</b>	111
			<b>4.5</b>	220
			<b>5.7</b>	400

# Future and Ongoing Work

Repeat the experiment with germanium (Takuhiro Fujiie)

Higher-order structure factors

Valuable to crystal dynamics, neutron charge radius, and fifth force searches

Global analysis of charge radius measurements for better separation of atomic and fifth force form factors (similar to Zimmer & Kaiser 2006)

Neutron interferometer with position-sensitive detector

Pendellosung on a pulsed Beam (Itoh et al 2018)

Pendellosung as a function of temperature

# Thank You!



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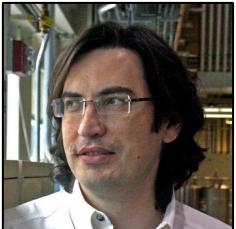
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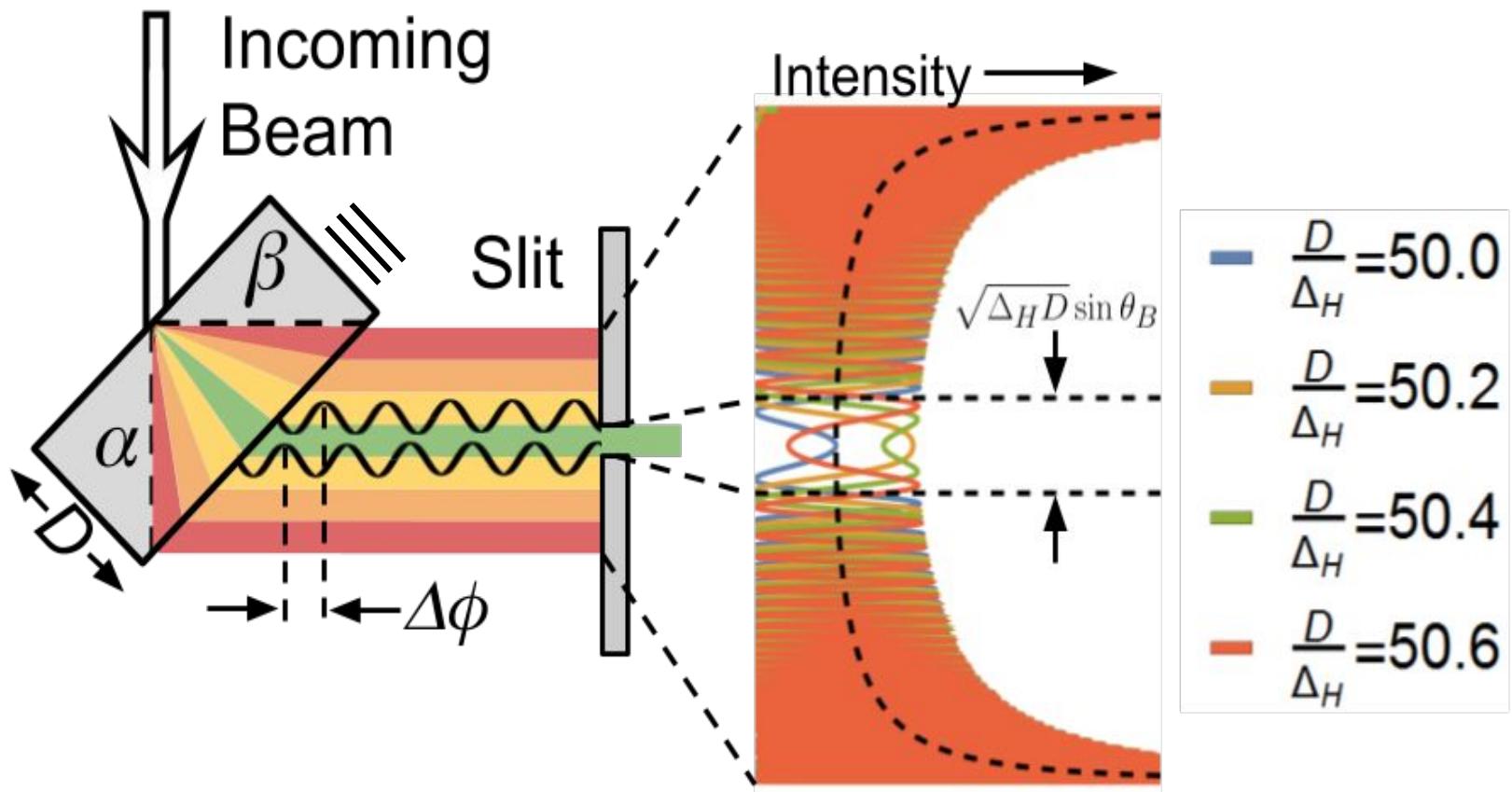
Special Thanks:  
Sam Werner



# Borrmann Fan

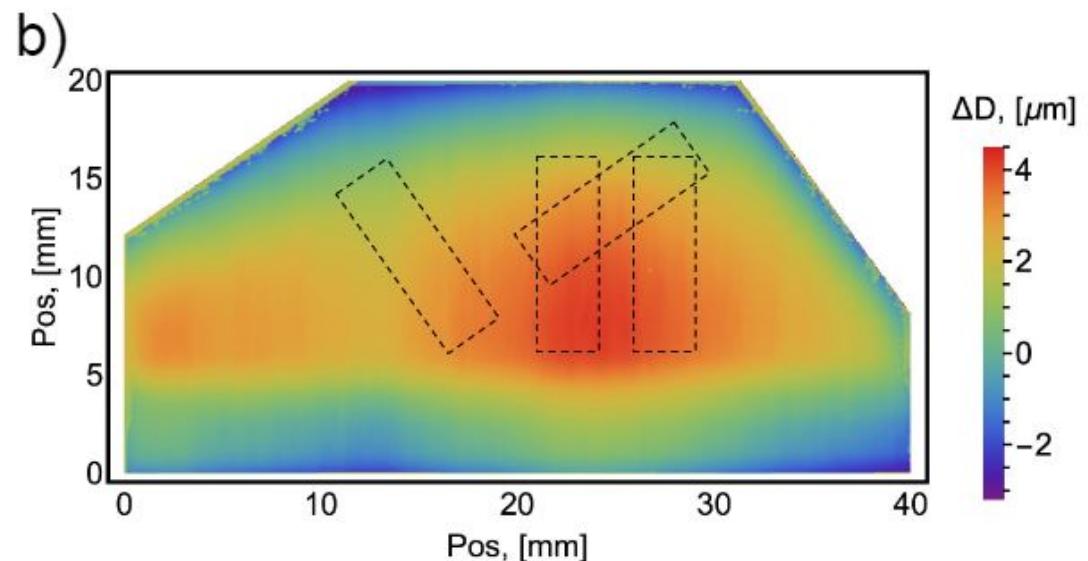
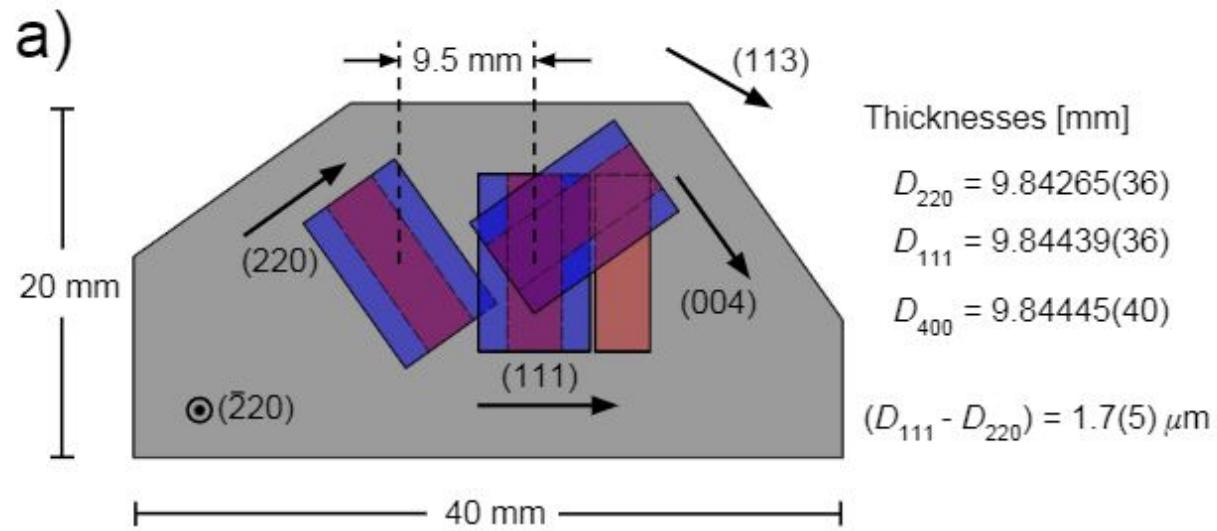
Edges of the Borrmann fan are detuned from the exact Bragg condition

Pendellosung must be measured at the center of the Borrmann fan



# Beam Profiles

Systematic uncertainty associated with different interrogation volumes of the neutron interferometer and pendellosung beam



# Pendellosung

Length domain

Two state system -  
momentum directions

Bragg misalignment

$$\vec{K} \cdot \vec{H} = \left( -\frac{1}{2} \vec{H} + \delta \vec{K} \right) \cdot \vec{H}$$

Pendellosung length

Strength of potential

$$\Delta_H = \pi \frac{H \cot \theta_B}{v_H}$$

# Time Sinusoidal (NMR)

Time domain

Two state system -  
energy levels

Resonance condition

$$\omega = \frac{E_2 - E_1}{\hbar} + \delta\omega$$

Rabi flopping

Strength of potential

# Anharmonic Contribution

The “nice” functional for of the DWF assumes a harmonic lattice potential

The different atoms with opposite bonding parities see opposite cubic contribution to interatomic potential

Cubic contribution is required for thermal expansion

Silicon’s negative coefficient of thermal expansion at low temperatures makes it especially interesting

$$\mathcal{H} = \frac{p_i^\alpha p_i^\alpha}{2m_i} + \frac{1}{2} \Phi_{ij}^{\alpha\beta} u_i^\alpha u_j^\beta + \frac{1}{3!} \Psi_{ijk}^{\alpha\beta\gamma} u_i^\alpha u_j^\beta u_k^\gamma$$

# Cubic Terms in Silicon

$$\left| \left\langle \sum_v e^{i\vec{H} \cdot \vec{x}_v} \right\rangle \right| = \begin{cases} \sqrt{32}e^{-W} [1 - T_a(hkl)], & h + k + l = 4n - 1 \\ & h, k, l \in \text{Odd} \\ \sqrt{32}e^{-W} [1 + T_a(hkl)], & h + k + l = 4n + 1 \\ & h, k, l \in \text{Odd} \\ 8e^{-W}, & h + k + l = 4n \\ & h, k, l \in \text{Even} \\ 8e^{-W}T_a(hkl), & h + k + l = 4n \pm 2 \\ & h, k, l \in \text{Even} \end{cases}$$

$$T_a(hkl) = \frac{1}{6} \left\langle (\vec{H} \cdot \vec{u})^3 \right\rangle = \left( \frac{B}{4\pi a} \right)^3 \frac{\beta}{k_B T} hkl \simeq (3 \times 10^{-5}) hkl$$

$$\beta = \Psi_{iii}^{xyz}$$

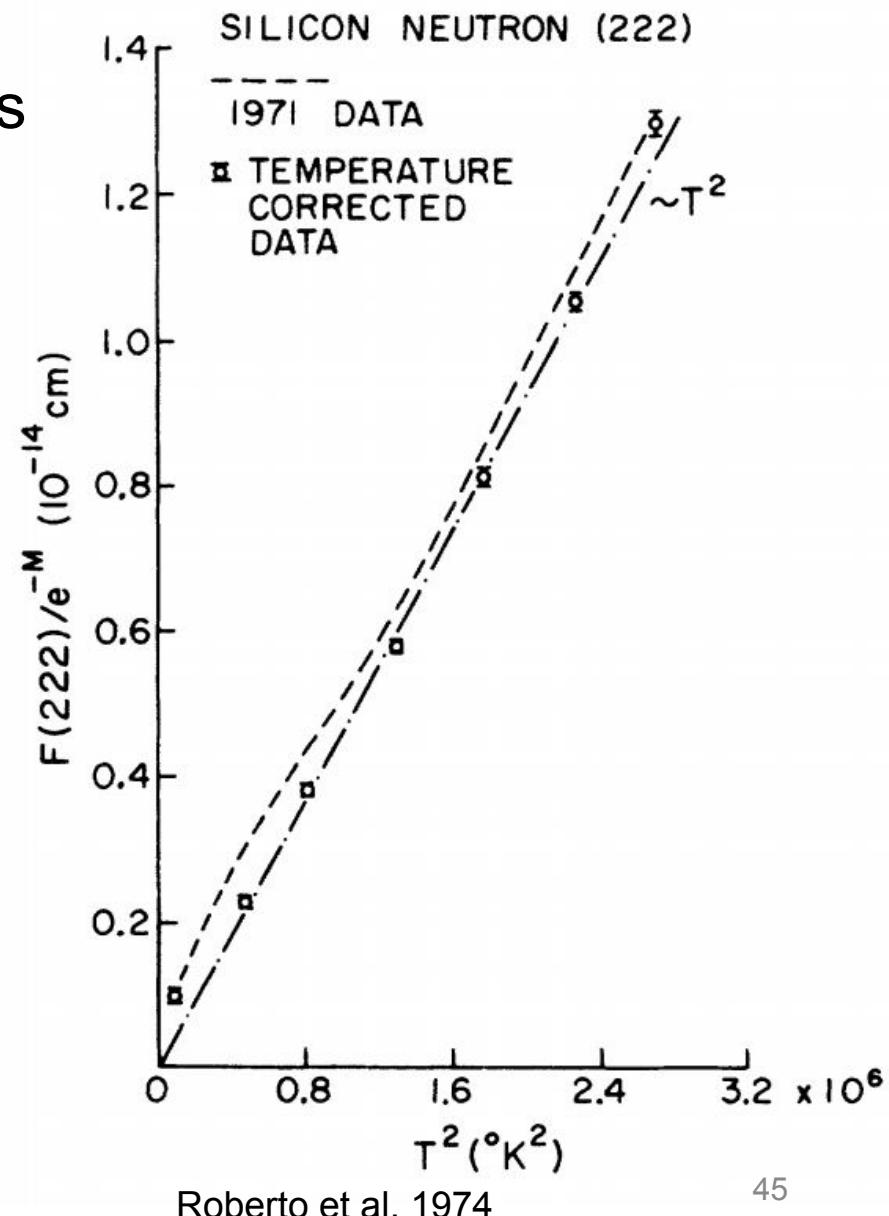
# Forbidden Reflections

One class of forbidden reflections  
is nonzero due to

Bonding Electrons

Anharmonic atomic motion

Measured using absolute  
reflectivity as a function of  
temperature



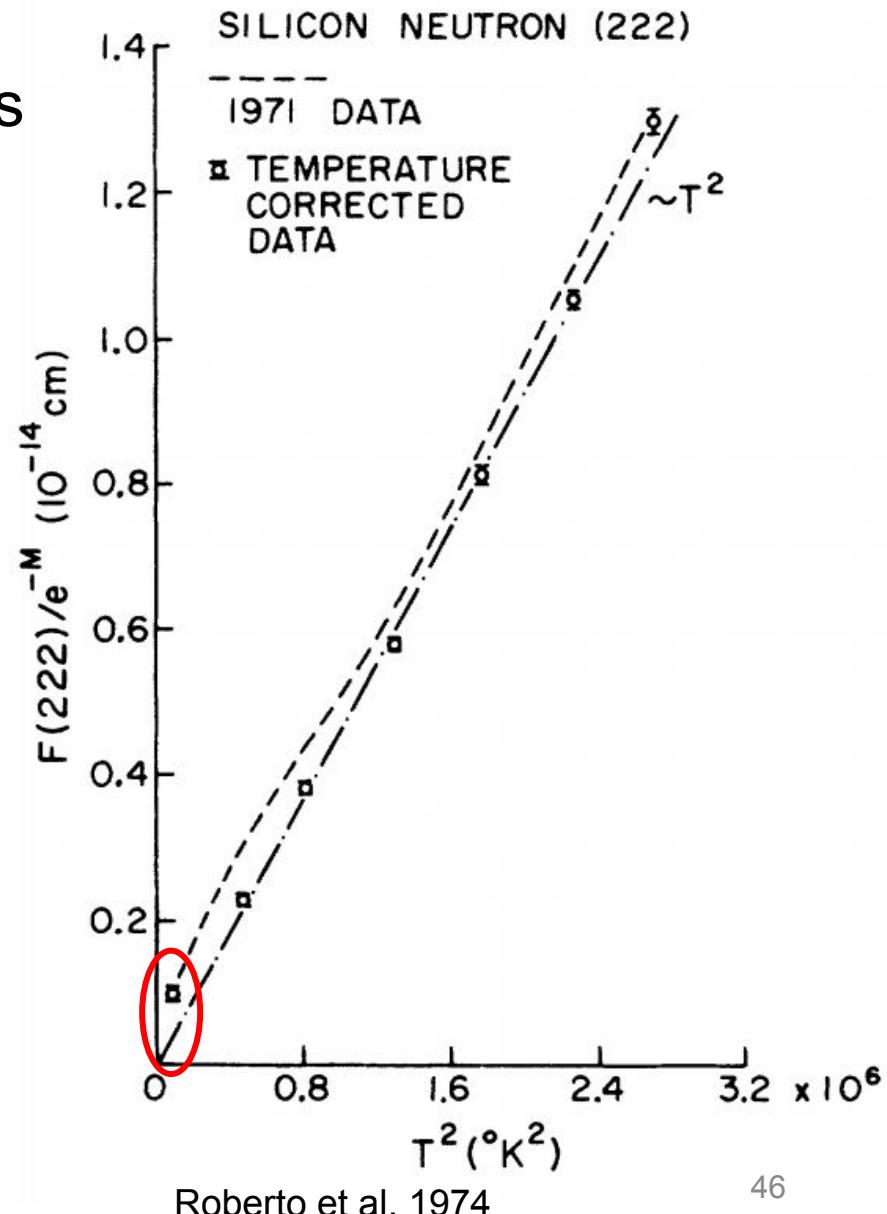
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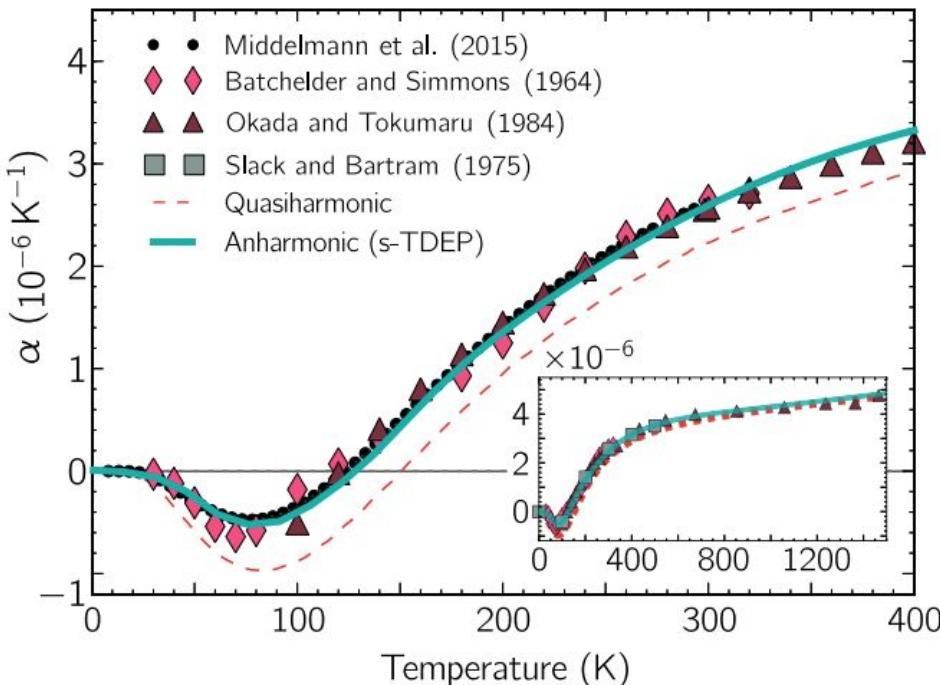


# Nuclear quantum effect with pure anharmonicity and the anomalous thermal expansion of silicon

D. S. Kim<sup>a,1,2</sup>, O. Hellman<sup>a,1</sup>, J. Herriman<sup>a</sup>, H. L. Smith<sup>a</sup>, J. Y. Y. Lin<sup>b</sup>, N. Shulumba<sup>c</sup>, J. L. Niedziela<sup>d</sup>, C. W. Li<sup>e</sup>, D. L. Abernathy<sup>f</sup>, and B. Fultz<sup>a,2</sup>

<sup>a</sup>Department of Applied Physics and Materials Science, California Institute of Technology, Pasadena, CA 91125; <sup>b</sup>Neutron Data Analysis and Visualization Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831; <sup>c</sup>Department of Mechanical and Civil Engineering, California Institute of Technology, Pasadena, CA 91125; <sup>d</sup>Instrument and Source Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831; <sup>e</sup>Department of Mechanical Engineering, University of California, Riverside, CA 92521; and <sup>f</sup>Quantum Condensed Matter Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831

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## Significance

Silicon has a peculiar negative thermal expansion at low temperature. This behavior has been understood with a “quasi-harmonic” theory where low-energy phonons decrease in frequency with volume contraction. We report inelastic neutron scattering measurements of phonon dispersions over a wide range of temperatures. These measurements cast doubt upon quasiharmonic theory, which predicts the wrong sign for most phonon shifts with temperature. Fully anharmonic ab initio calculations correctly predict the phonon shifts and thermal expansion. Crystal structure, anharmonicity, and nuclear quantum effects all play important roles in the thermal expansion of silicon, and a simple mechanical explanation is inappropriate. The quantum effect of nuclear vibrations is also expected to be important for thermophysical properties of many materials.