Neutron Scattering
with Examples from Cuprate Superconductors
Part 1

John Tranquada

BROOKHAVEN NATIONAL LABORATORY

Quantum Science Summer School
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U.S. DEPARTMENT OF ENERGY | Office of Science

CES center for emergent superconductivity

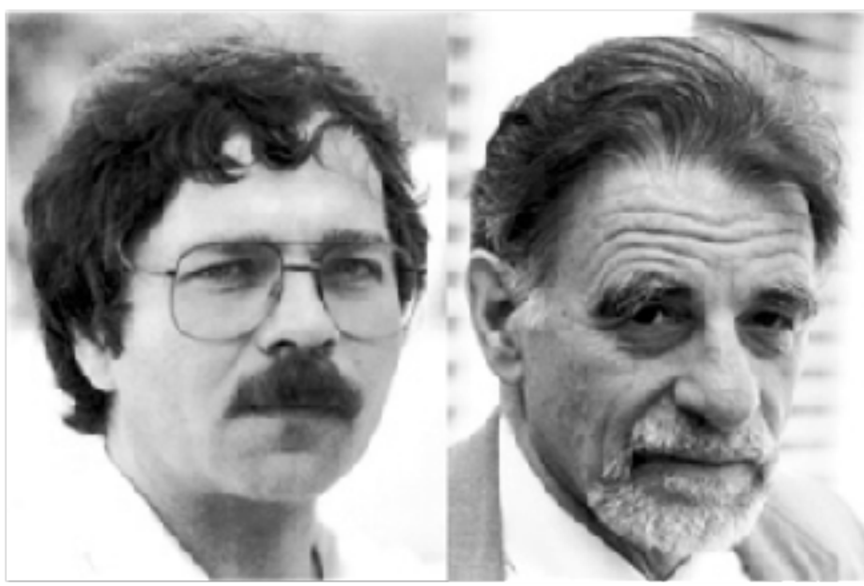
> 30 years since discovery of high T_c superconductivity

Z. Phys. B – Condensed Matter 64, 189–193 (1986)

Possible High T_c Superconductivity in the Ba – La – Cu – O System

J.G. Bednorz and K.A. Müller
IBM Zürich Research Laboratory, Rüschlikon, Switzerland

Received April 17, 1986



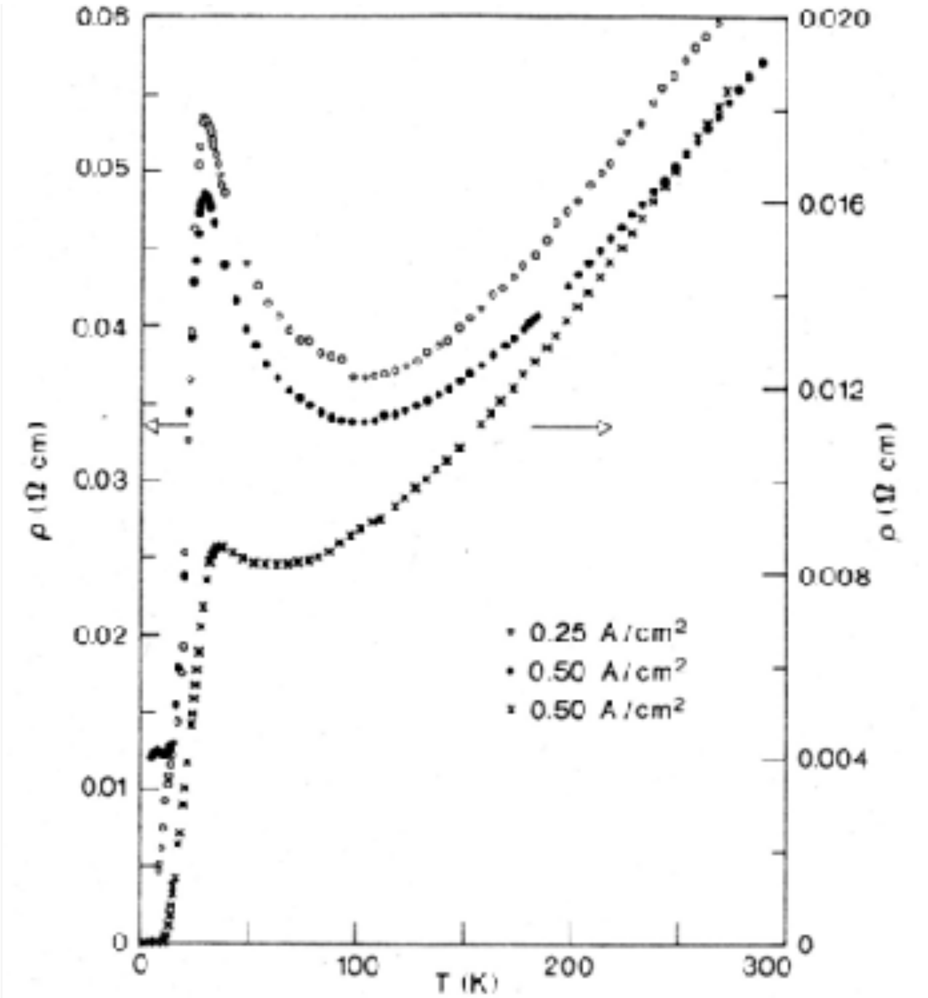


Fig. 1. Temperature dependence of resistivity in $Ba_xLa_{5-x}Cu_5O_{5(3-y)}$ for samples with $x(Ba) = 1$ (upper curves, left scale) and $x(Ba) = 0.75$ (lower curve, right scale). The first two cases also show the influence of current density

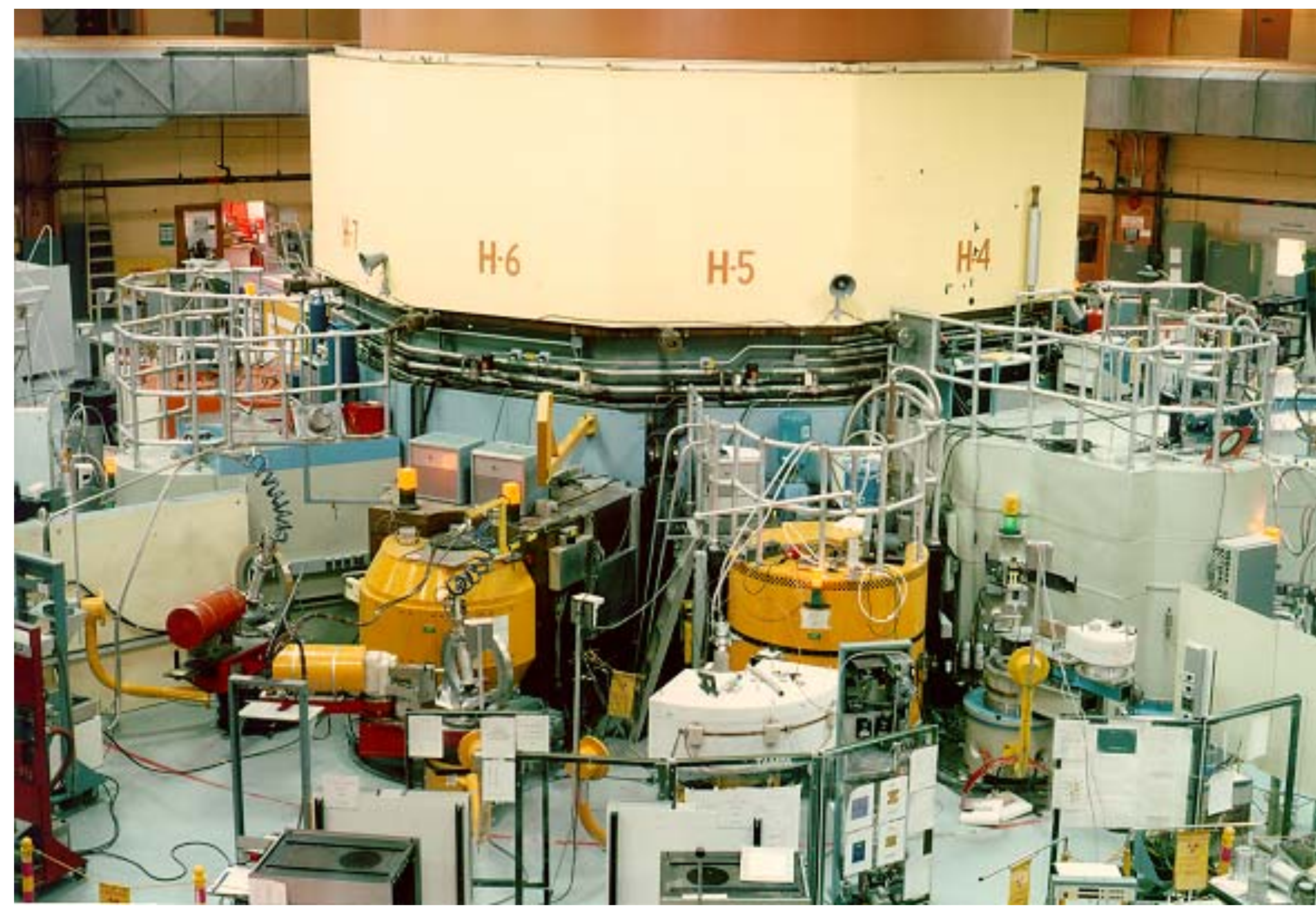
Superconducting phase:

$La_{2-x}Ba_xCuO_4$

Zürich oxide

High Flux Beam Reactor (BNL)

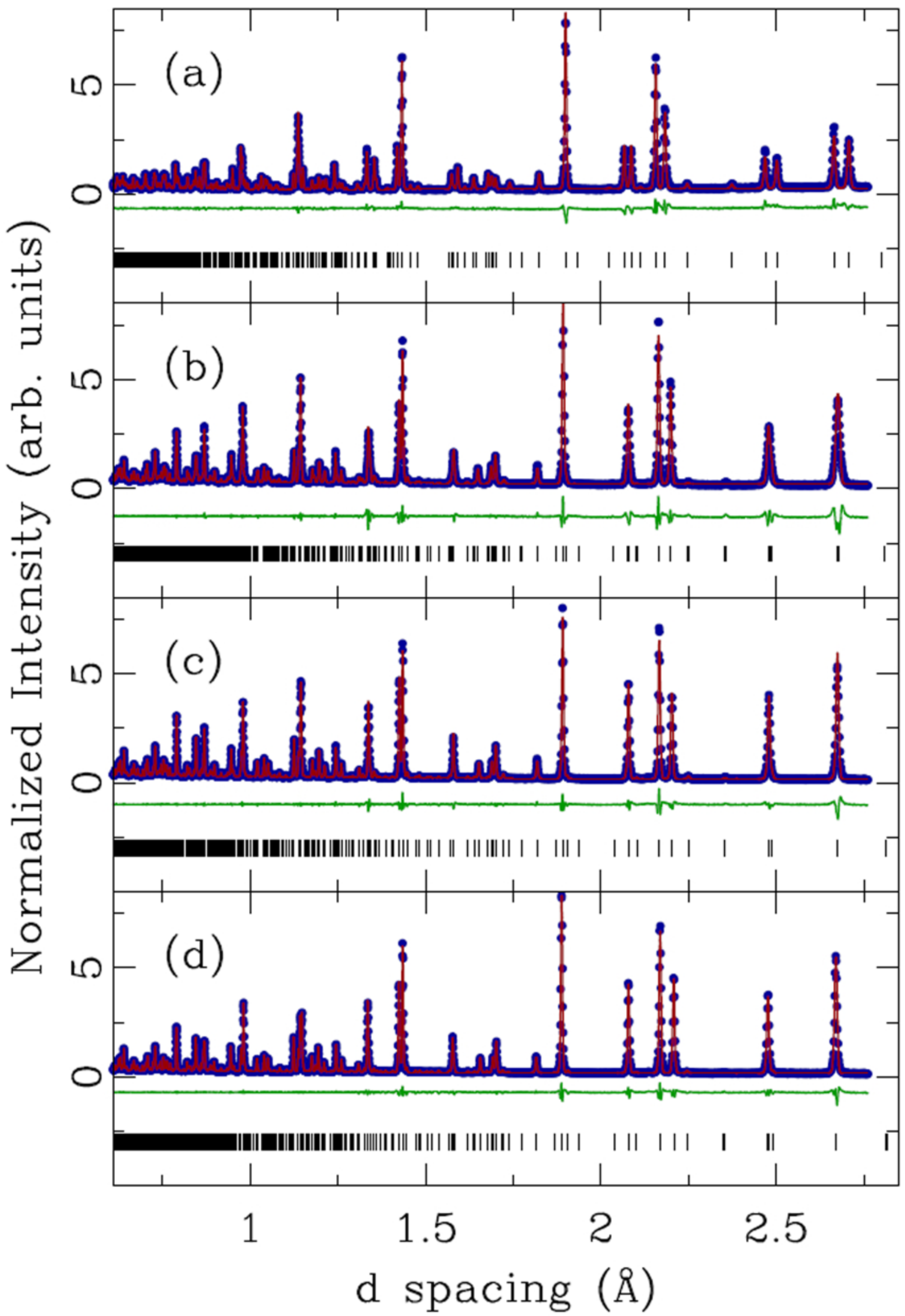
1965 - HFBR first goes critical

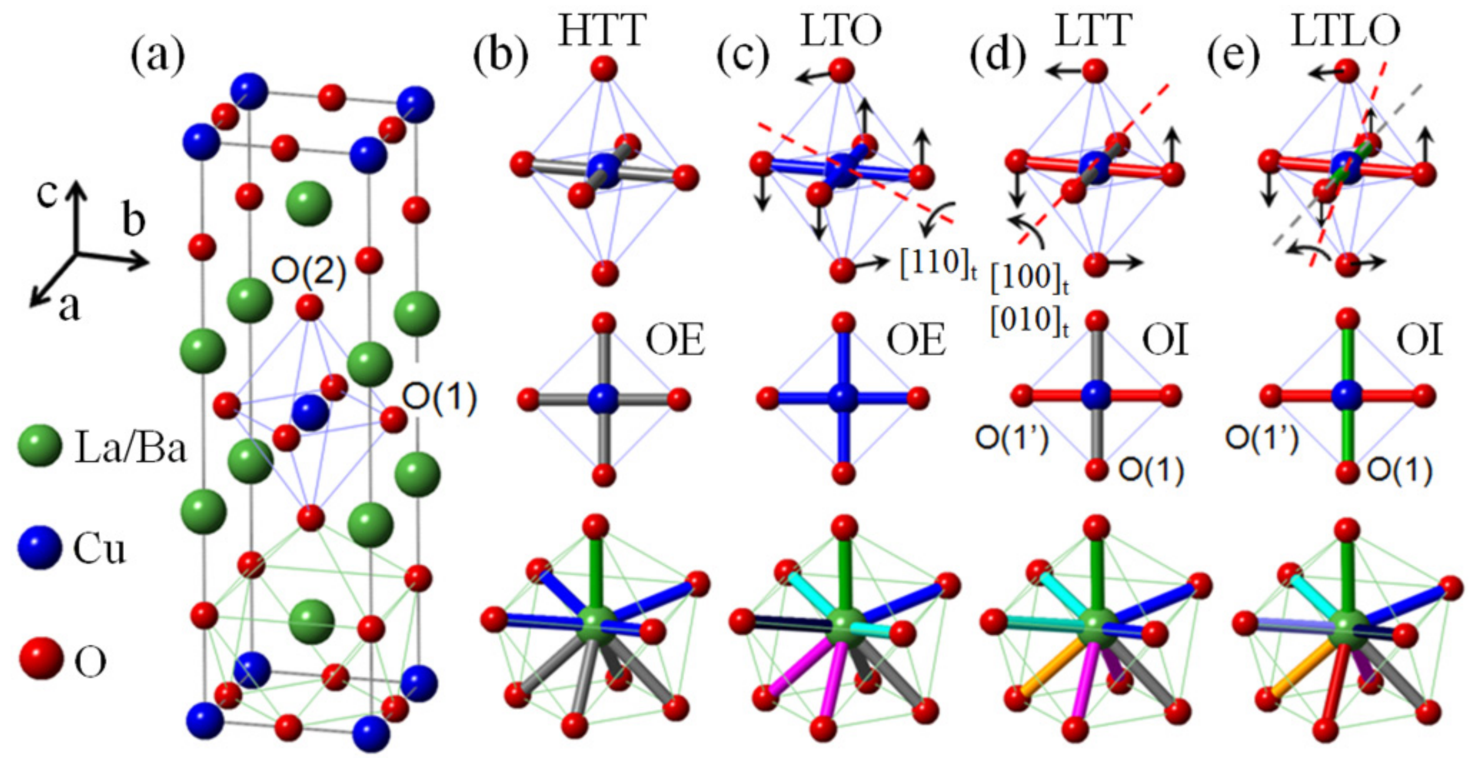


1999 - HFBR officially closed

Crystal structure of $La_{2-x}Ba_xCuO_4$

Neutron Powder Diffraction





Bozin *et al*., PRB **91**, 054521 (2015).

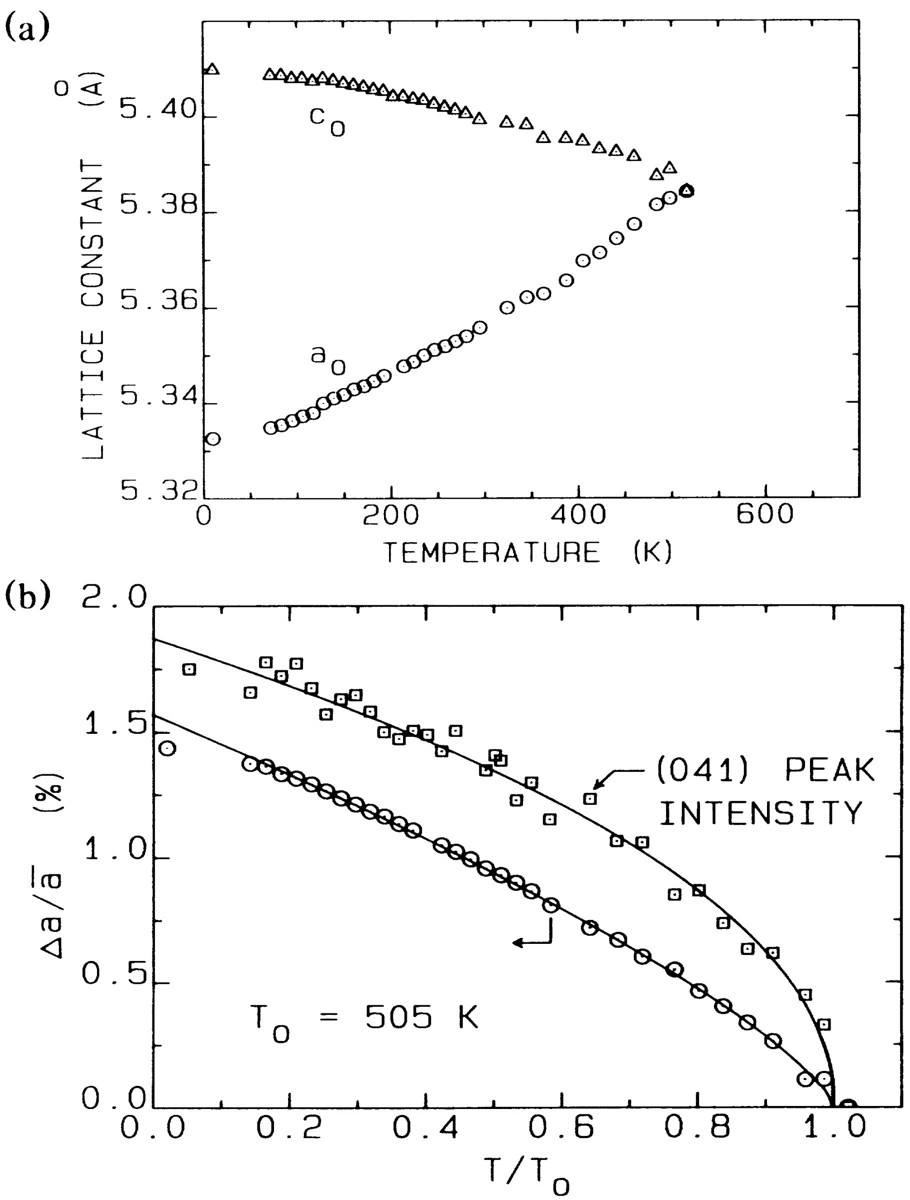
"Lattice instability and high-T_c superconductivity in $La_{2-x}Ba_xCuO_4$"
J. D. Jorgensen *et al*., Phys. Rev. Lett. 58, 1024 (1987).

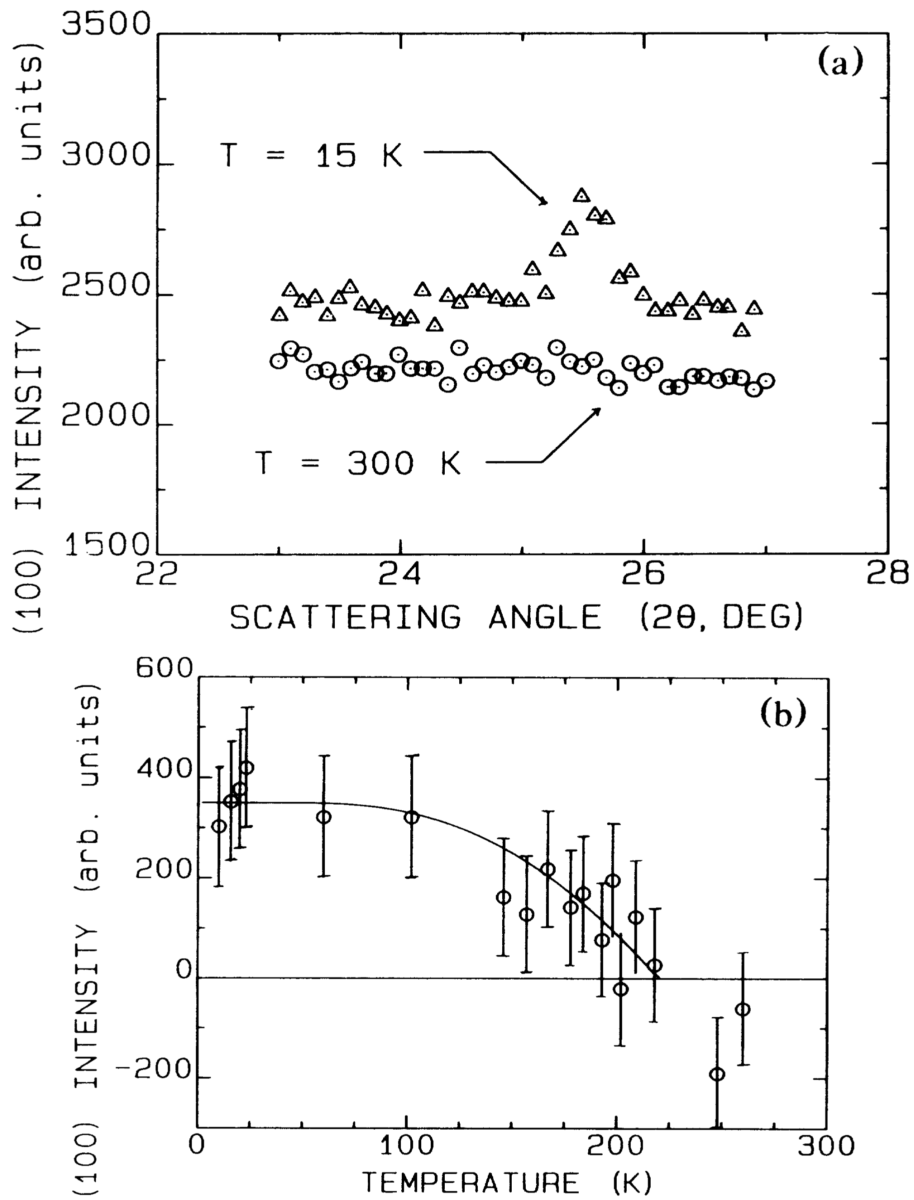
"Structural phase transformations and superconductivity in $La_{2-x}Ba_xCuO_4$"
J. D. Axe *et al*., Phys. Rev. Lett. 62, 2751 (1989).

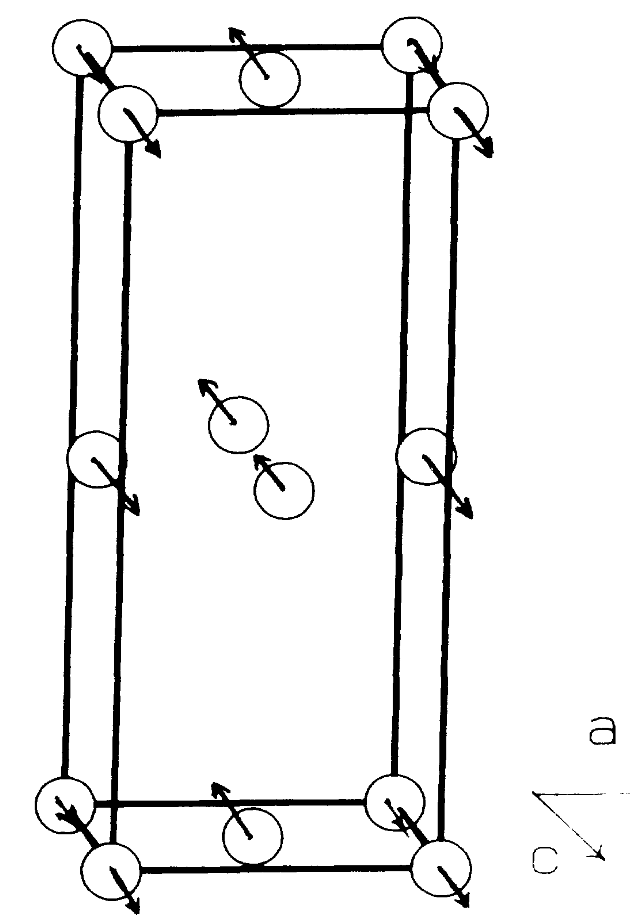
Antiferromagnetism in La_2CuO_{4-y}

D. Vaknin,[a] S. K. Sinha, D. E. Moncton, D. C. Johnston, J. M. Newsam, C. R. Safinya, and H. E. King, Jr.
Corporate Research Laboratories, Exxon Research and Engineering Company, Annandale, New Jersey 08801

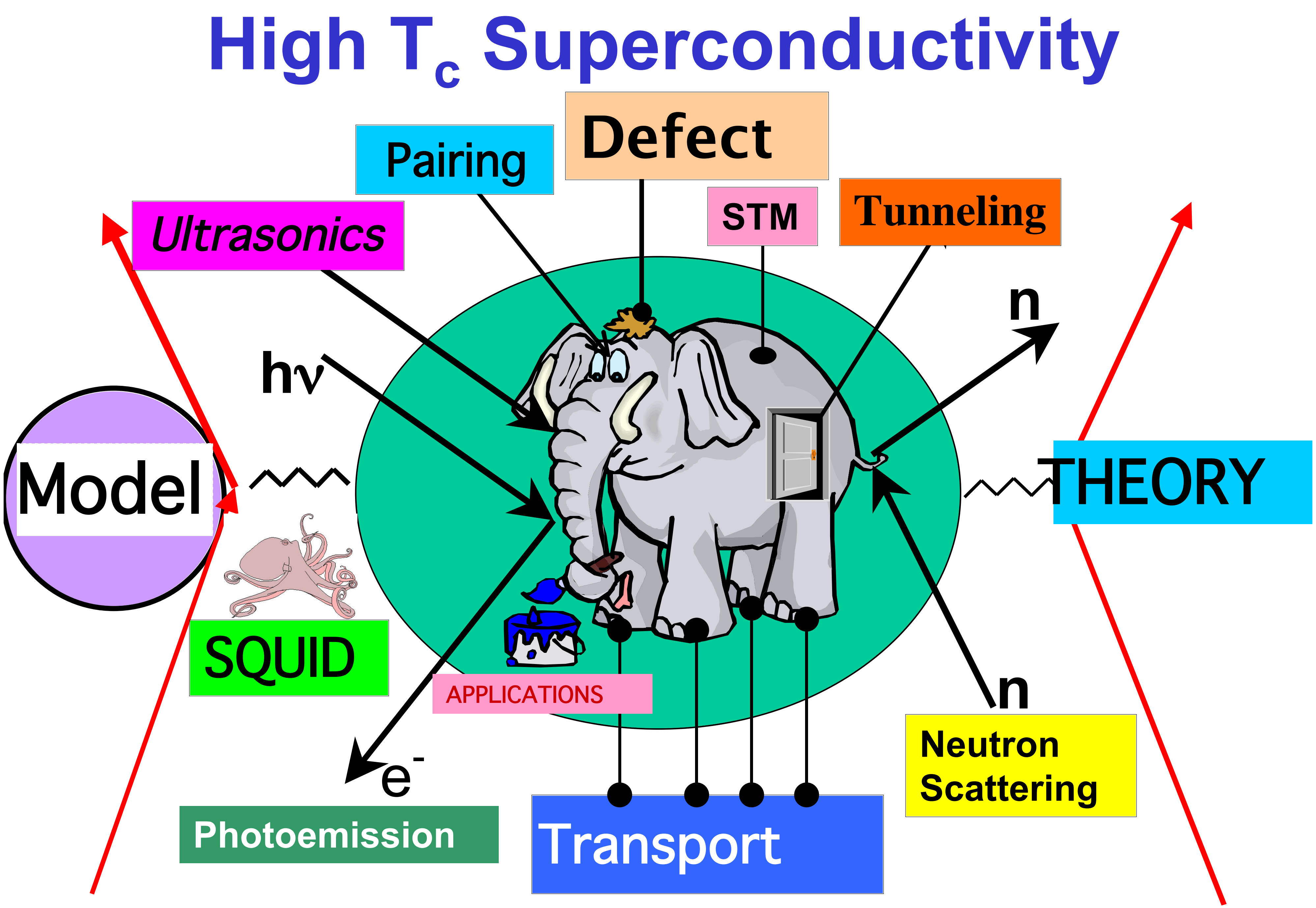
(Received 4 May 1987)







The blind physicists and the superconductor



I. K. Schuller *et al*., DOE report (2002)

Properties of the Neutron

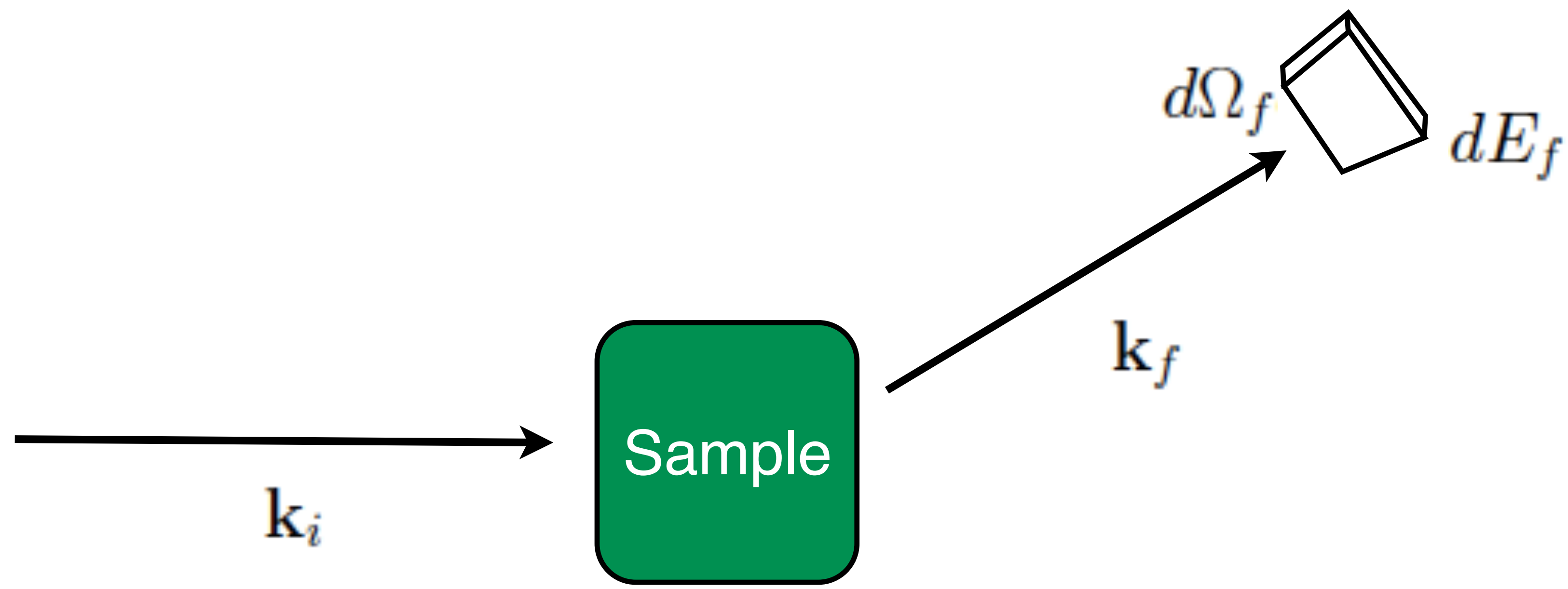
mass	$m_n = 1.009$ amu
	$\approx m_p \approx 1838\ m_e$
spin	1/2
magnetic moment	$1.913\ \mu_N$
	$0.001\ \mu_B$
charge	0

Conversions and units

Table 2: Wavelength, frequency, velocity and energy relationships for neutrons.

Quantity	Relationship	Value at $E = 10$ meV
Energy	$E[\mathrm{meV}] = 2.072k^2[\text{Å}^{-1}]$	10 meV
Wavelength	$\lambda[\text{Å}] = 9.044/\sqrt{E[\mathrm{meV}]}$	2.86 Å
Wavevector	$k[\text{Å}^{-1}] = 2\pi/\lambda[\text{Å}]$	2.20 Å^{-1}
Frequency	$\nu[\mathrm{THz}] = 0.2418E[\mathrm{meV}]$	2.418 THz
Wavenumber	$\nu[\mathrm{cm}^{-1}] = \nu[\mathrm{Hz}]/(2.998 \times 10^{10}\ \mathrm{cm/sec})$	80.65 cm^{-1}
Velocity	$v[\mathrm{km/sec}] = 0.6302k[\text{Å}^{-1}]$	1.38 km/sec
Temperature	$T[\mathrm{K}] = 11.605E[\mathrm{meV}]$	116.05 K

Neutron scattering



$$k = \frac{2\pi}{\lambda}$$

$d\Omega_f$ = differential of solid angle for $\mathbf{k}_f$

$$E = \frac{\hbar^2 k^2}{2m_\mathrm{n}}$$

dE_f = energies between E_f and $E_f + dE_f$

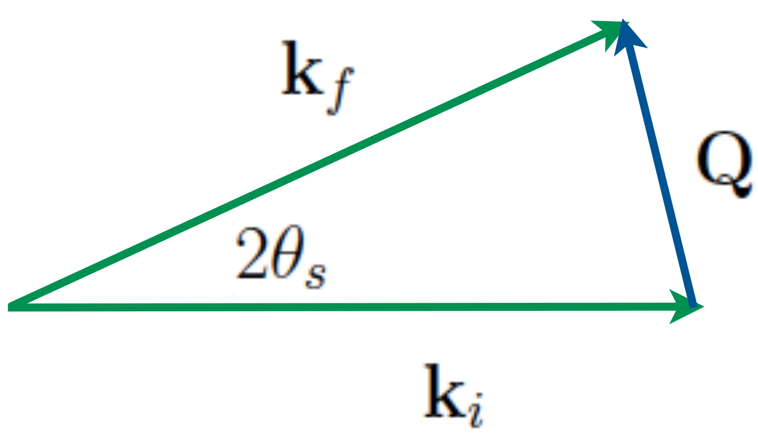
$\dfrac{d^2\sigma}{d\Omega_f dE_f}$ = probability of scattering into $d\Omega_f dE_f$

Momentum and energy transfer

$$\mathbf{Q} = \mathbf{k}_f - \mathbf{k}_i \qquad \text{(momentum conservation)}$$

$$|\mathbf{Q}| = k_i^2 + k_f^2 - 2k_i k_f \cos(2\theta_s)$$

$$\begin{aligned} \hbar\omega &= E_i - E_f \qquad \text{(energy conservation)} \\ &= \frac{\hbar^2}{2m_\mathrm{n}}(k_i^2 - k_f^2) \end{aligned}$$



Differential cross section

Fermi's golden rule:

$$\left.\frac{d^2\sigma}{d\Omega_f dE_f}\right|_{\lambda_i\to\lambda_f} = \frac{k_f}{k_i}\left(\frac{m_\mathrm{n}}{2\pi\hbar^2}\right)^2 |\langle \mathbf{k}_f\lambda_f|V|\mathbf{k}_i\lambda_i\rangle|^2\delta(\hbar\omega + E_i - E_f)$$

Born approximation (treat neutrons as plane waves):

$$\langle \mathbf{k}_f\lambda_f|V|\mathbf{k}_i\lambda_i\rangle = V(\mathbf{Q})\Big\langle\lambda_f\Big|\sum_l e^{i\mathbf{Q}\cdot\mathbf{r}_l}\Big|\lambda_i\Big\rangle$$

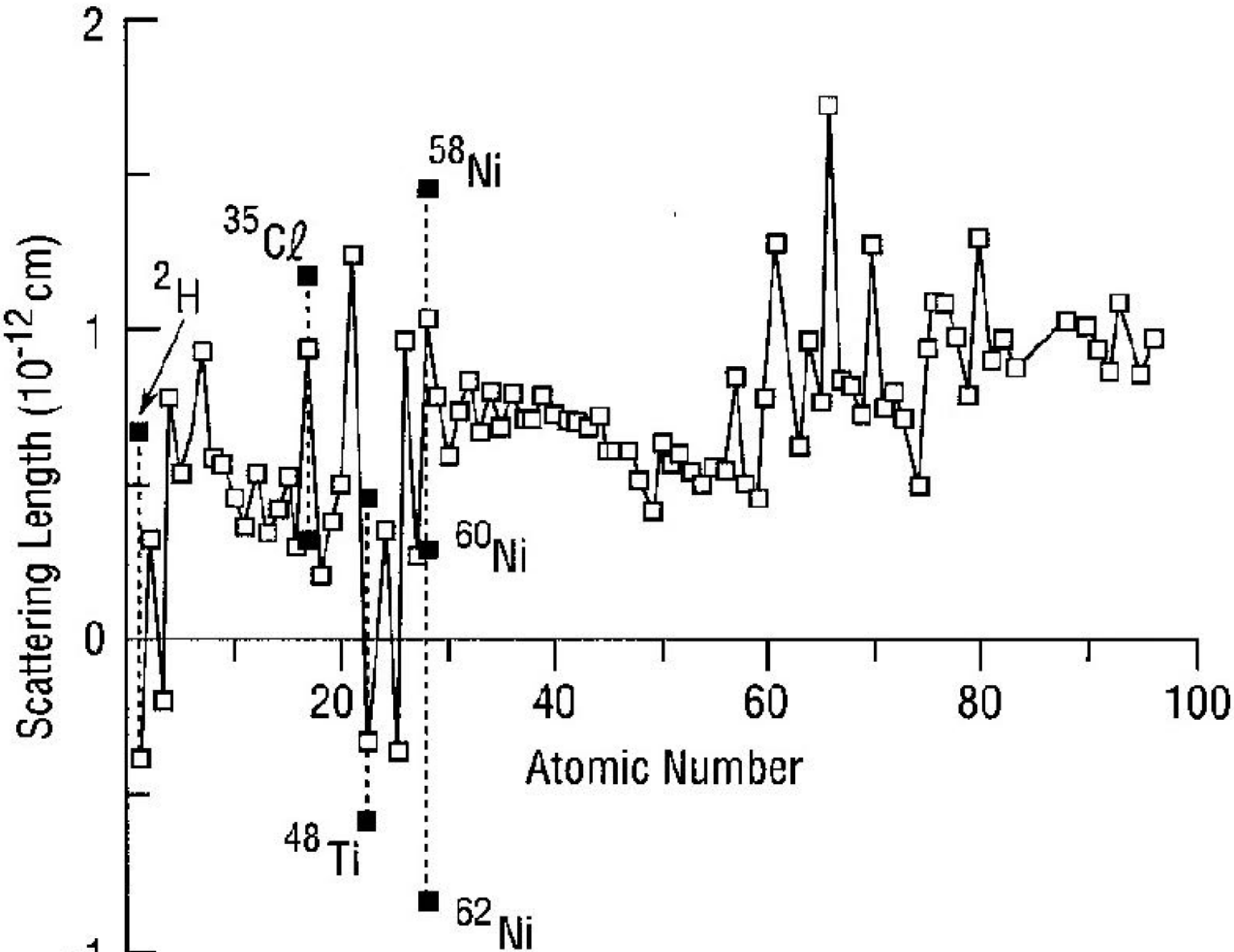
for a collection of atoms at positions $\mathbf{r}_l$

$$V(\mathbf{Q}) = \int d\mathbf{r}\, V(\mathbf{r})e^{i\mathbf{Q}\cdot\mathbf{r}}$$

Nuclear scattering:

$$V(\mathbf{Q}) = \frac{2\pi\hbar^2}{m_\mathrm{n}}b$$

Nuclear scattering lengths



Coherent and incoherent scattering

Distribution of nuclear isotopes (or nuclear spins) with scattering factors b_r and frequencies c_r

Coherent scattering from a collection of atoms depends on the average scattering length:

$$\bar{b} = \sum_r c_r b_r$$

$$\sigma_{\mathrm{coh}} = 4\pi \left(\bar{b}\right)^2$$

Total scattering:

$$\sigma_{\mathrm{scat}} = 4\pi \sum_r c_r b_r^2 = 4\pi \overline{b^2}$$

Incoherent scattering = difference between total and coherent scattering, corresponds to scattering from individual atoms:

$$\sigma_{\mathrm{inc}} = 4\pi \left(\overline{b^2} - \bar{b}^2\right) = 4\pi \overline{(b - \bar{b})^2}$$

$$b_{\mathrm{inc}} = \sqrt{\overline{b^2} - \bar{b}^2}$$

Examples of coherent vs. incoherent

Table 3: Neutron scattering lengths and cross sections.

Isotope	Natural Abundance (%)	b_{coh} (fm)	b_{inc} (fm)
H		$-3.7390(11)$	
^{1}H	99.985	$-3.7406(11)$	$25.274(9)$
^{2}H	0.015	$6.671(4)$	$4.04(3)$
^{3}H	(12.32 a)	$4.792(27)$	$-1.04(17)$
Be	100	$7.79(1)$	$0.12(3)$
B		$5.30(4) - 0.213(2)i$	
^{10}B	20	$-0.1(3) - 1.066(3)i$	$-4.7(3) + 1.231(3)i$
^{11}B	80	$6.65(4)$	$-1.3(2)$

Scattering lengths in $La_{2-x}(Ba,Sr)_xCuO_4$

Element	b_{coh} (fm)	Z
La	8.24	57
Ba	5.07	56
Sr	7.02	38
Cu	7.72	29
O	5.80	16

In transition-metal oxides, neutrons are more sensitive to oxygen than x-rays are.

Distribution of states in sample

Average over initial states, sum over final states:

$$\frac{d^2\sigma}{d\Omega_f dE_f} = \frac{k_f}{k_i} \sum_{\lambda_i,\lambda_f} P(\lambda_i) \left| \langle \lambda_f | \sum_l e^{i\mathbf{Q}\cdot\mathbf{r}_l} | \lambda_i \rangle \right|^2 \delta(\hbar\omega + E_i - E_f)$$

$P(\lambda_i)$ = distribution of initial states

Dynamical structure factor (Van Hove)

$$\frac{d^2\sigma}{d\Omega_f dE_f} = N \frac{k_f}{k_i} b^2 S(\mathbf{Q},\omega)$$

$$S(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar N} \sum_{ll'} \int_{-\infty}^{\infty} dt \, \langle e^{-i\mathbf{Q}\cdot\mathbf{r}_{l'}(0)} e^{i\mathbf{Q}\cdot\mathbf{r}_l(t)} \rangle \, e^{-i\omega t}$$

$$\langle \ldots \rangle = \sum_i P(\lambda_i) \ldots$$

Coherent elastic nuclear scattering

Time-averaged structure factor:

$$S_{\mathrm{coh}}(\mathbf{Q},\omega) = \delta(\hbar\omega)\frac{1}{N}\left\langle \sum_{ll'} e^{i\mathbf{Q}\cdot(\mathbf{r}_l - \mathbf{r}_{l'})} \right\rangle$$

Single crystal with a Bravais lattice (single atom per unit cell):

$$S_{\mathrm{coh}}(\mathbf{Q},\omega) = \delta(\hbar\omega)\frac{(2\pi)^3}{v_0}\sum_{\mathbf{G}} \delta(\mathbf{Q}-\mathbf{G})$$

$\mathbf{G}$ = reciprocal lattice wave vector

v_0 = volume per unit cell

Coherent elastic cross section:

$$\left.\frac{d\sigma}{d\Omega}\right|_{\mathrm{coh}}^{\mathrm{el}} = N\frac{(2\pi)^3}{v_0}\left(\bar{b}\right)^2\sum_{\mathbf{G}} \delta(\mathbf{Q}-\mathbf{G})$$

Generalizations

Impact of lattice vibrations:

$$\langle e^{i\mathbf{Q}\cdot\mathbf{r}} \rangle = e^{-2W} \qquad \text{Debye-Waller factor}$$

$$W = \frac{1}{2}\left\langle (\mathbf{Q}\cdot\mathbf{u})^2 \right\rangle$$

Average is over atomic displacements $\mathbf{u}$

Multiple atoms per unit cell:

$$\left.\frac{d\sigma}{d\Omega}\right|_c^{el} = N\frac{(2\pi)^3}{v_0}\sum_{\mathbf{G}}\delta(\mathbf{Q}-\mathbf{G})\left|F_N(\mathbf{G})\right|^2$$

$$F_N(\mathbf{G}) = \sum_j \bar{b}_j e^{i\mathbf{G}\cdot\mathbf{d}_j} e^{-W_j} \qquad \text{Nuclear structure factor}$$

Neutron Sources

- Neutrons live only in nuclei
  - Binding energy ~ 10 MeV

- Free neutrons obtained by:
  - Fission at a reactor source (steady state)
  - Spallation with a proton accelerator (pulsed, $\Delta t \sim 1\ \mu s$)

- Neutrons slowed with a moderator
  - Scattering from H (typically in H_2O, H_2, or CH_4)

- Neutron energy distribution ~ $e^{-E/kT}$
  - kT ~ 20 K ~ 2 meV   cold neutrons
  - kT ~ 300 K ~ 26 meV   thermal neutrons
  - 100 - 1000 meV   epithermal neutrons

Major Neutron User Facilities

Reactors

HFIR, Oak Ridge, TN

NCNR, Gaithersburg, MD

ILL, Grenoble, France

FRM-II, Munich, Germany

OPAL, Lucas Heights, Australia

JRR-3, Tokai, Japan (2020)

Spallation sources

SNS, Oak Ridge, TN

ISIS, Oxfordshire, UK

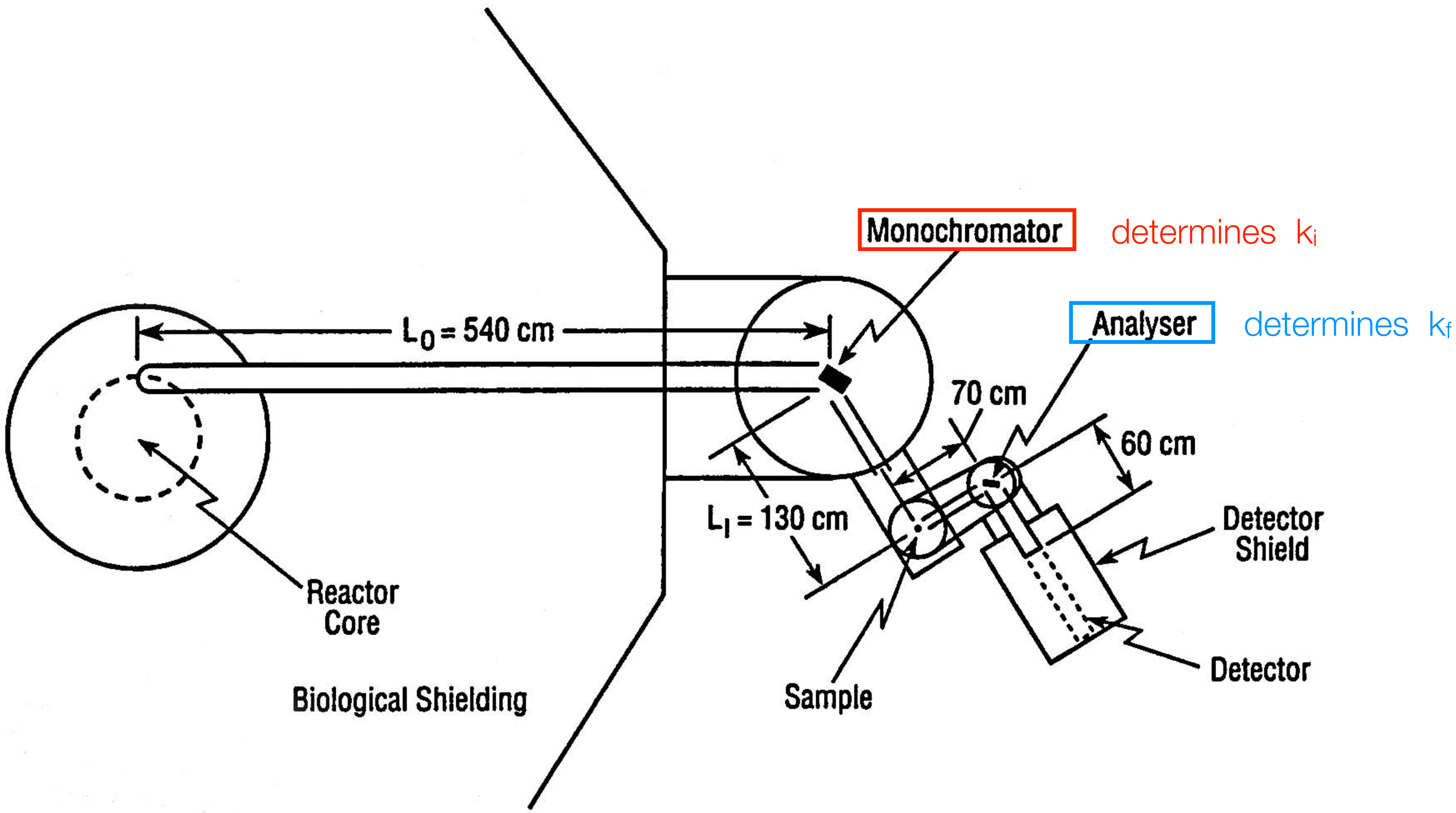
J-PARC, Tokai, Japan

SINQ, Villigen, Switzerland

CSNS, Dongguan, China

ESS, Lund, Sweden (2023)

Triple-axis spectrometer

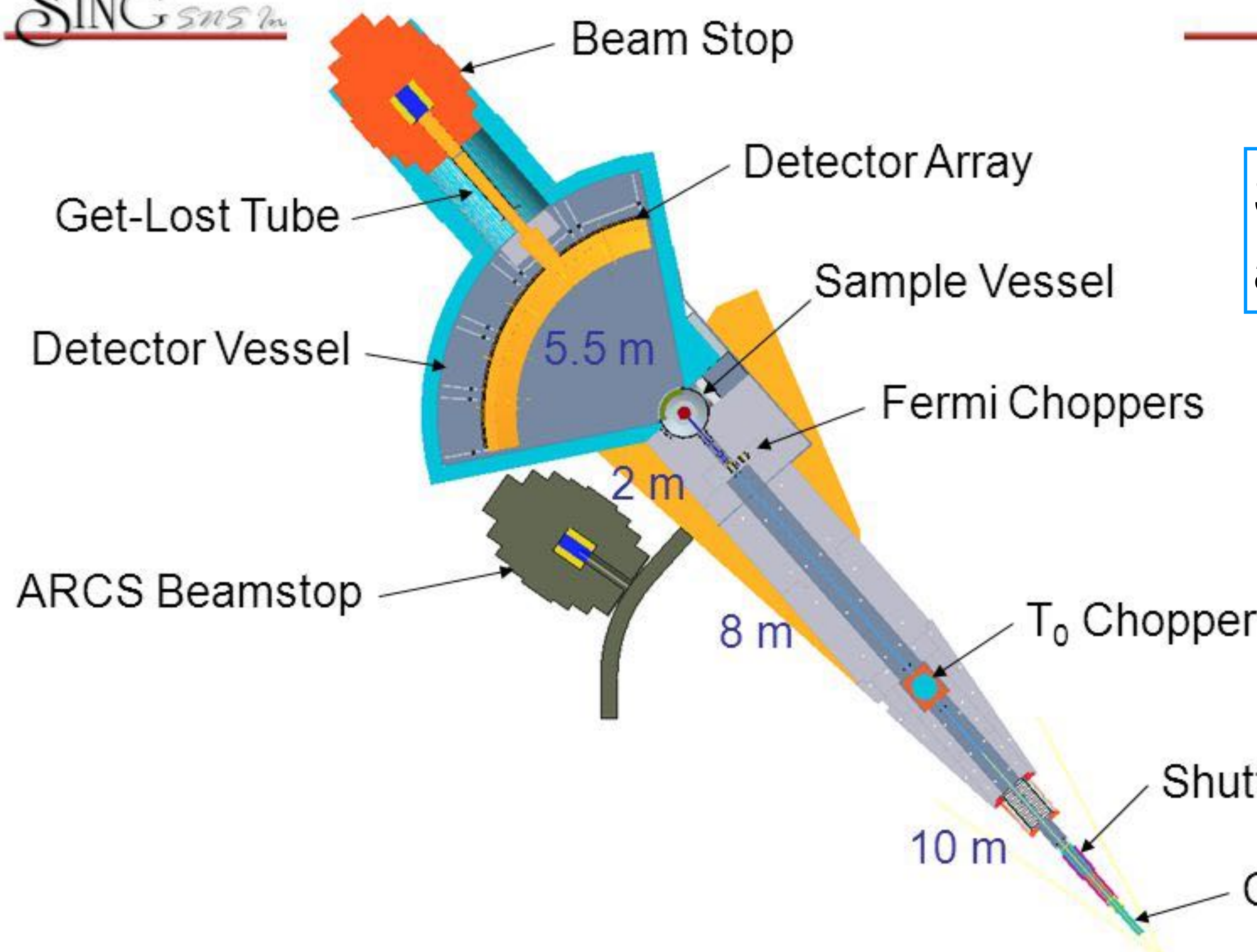


Advantage: very flexible, can tune resolution,
can handle many different sample environments

Disadvantage: not optimized for any particular experiment

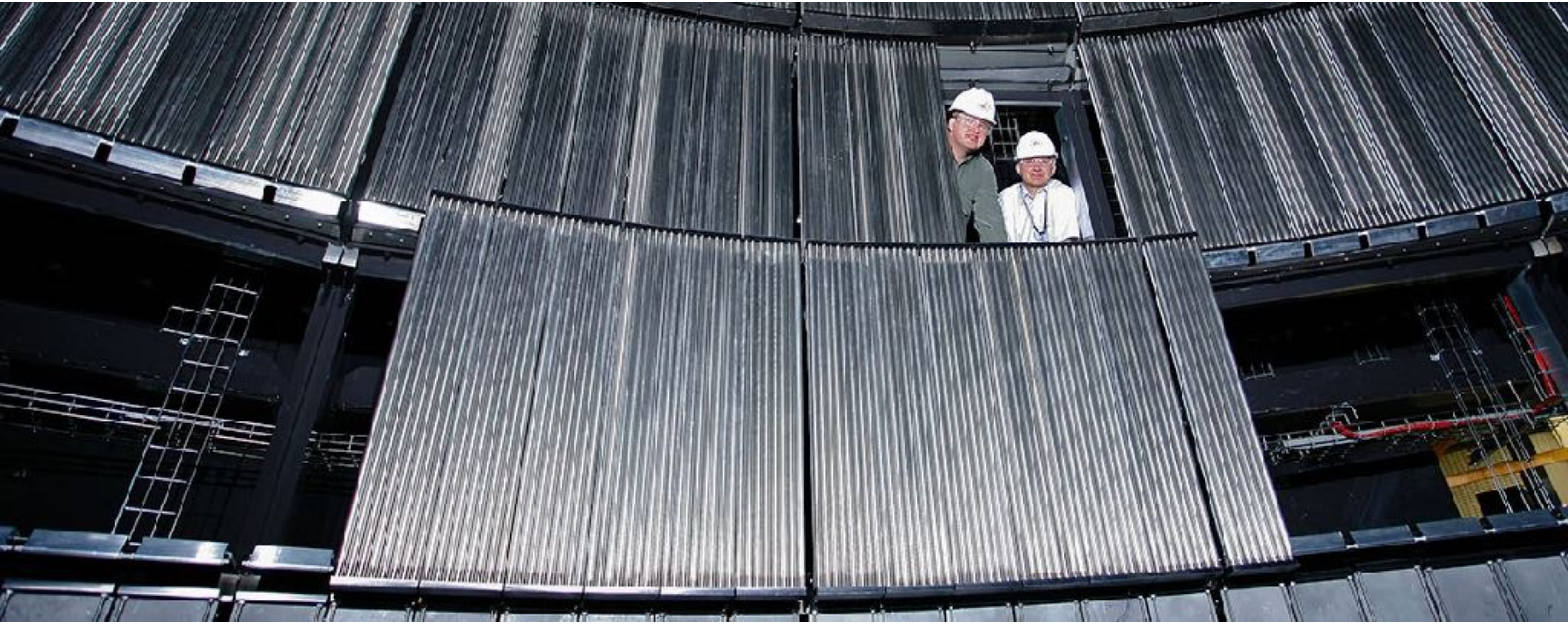
BT7 at NIST Center for Neutron Research

Time-of-Flight Spectrometer (Direct Geometry)



SEQUOIA at SNS

Position (and Time)-Sensitive Neutron Detectors



Detector installation at SEQUOIA

Scattering possibilities

- Instrument defines k_i and k_f
- $k_i = k_f$
  - Elastic scattering
    - Bragg peaks - from ordered lattice
    - Diffuse scattering - from disorder
- $k_i \neq k_f$
  - Inelastic scattering
    - Sharp excitations, phonons or magnons
    - Diffuse scattering

Coherent inelastic scattering

Detailed balance:

$$S(-\mathbf{Q}, -\omega) = e^{-\hbar\omega/k_{\mathrm{B}}T} S(\mathbf{Q}, \omega)$$

Fluctuation-dissipation theorem gives:

$$S(\mathbf{Q}, \omega) = \frac{\chi''(\mathbf{Q}, \omega)}{1 - e^{-\hbar\omega/k_B T}}$$

Dynamic susceptibility:

$$\chi(\mathbf{Q}, \omega) = \chi'(\mathbf{Q}, \omega) + i\chi''(\mathbf{Q}, \omega)$$

$$\chi''(\mathbf{Q}, -\omega) = -\chi''(\mathbf{Q}, \omega)$$

In an ordered state, $\chi''(\mathbf{Q}, \omega)$ for phonons (magnons) will have weak temperature dependence.

Phonons

Phonons have a dispersion $\omega_{\mathbf{q}s}$ where s labels the phonon modes and $\mathbf{q}$ is defined relative to a reciprocal lattice vector

$$\mathbf{Q} = \mathbf{G} + \mathbf{q}$$

There are always 3 acoustic modes (2 with transverse polarization, 1 with longitudinal polarization). For *n* atoms per unit cell, there will be 3*n* - 3 optical modes.

Acoustic modes at small q

$$\omega_{\mathbf{q}s} \approx v_s q$$

Phonon dispersions are the same in every Brillouin zone but the intensities can vary with $\mathbf{Q}$

Phonon susceptibility

$$\chi''(\mathbf{Q},\omega) = \frac{1}{2}\frac{(2\pi)^3}{v_0}\sum_{\mathbf{G},\mathbf{q}}\delta(\mathbf{Q}-\mathbf{q}-\mathbf{G})\sum_s \frac{1}{\omega_{\mathbf{q}s}}|\mathcal{F}(\mathbf{Q})|^2$$
$$\times\left[\delta\left(\omega-\omega_{\mathbf{q}s}\right)-\delta\left(\omega+\omega_{\mathbf{q}s}\right)\right]$$

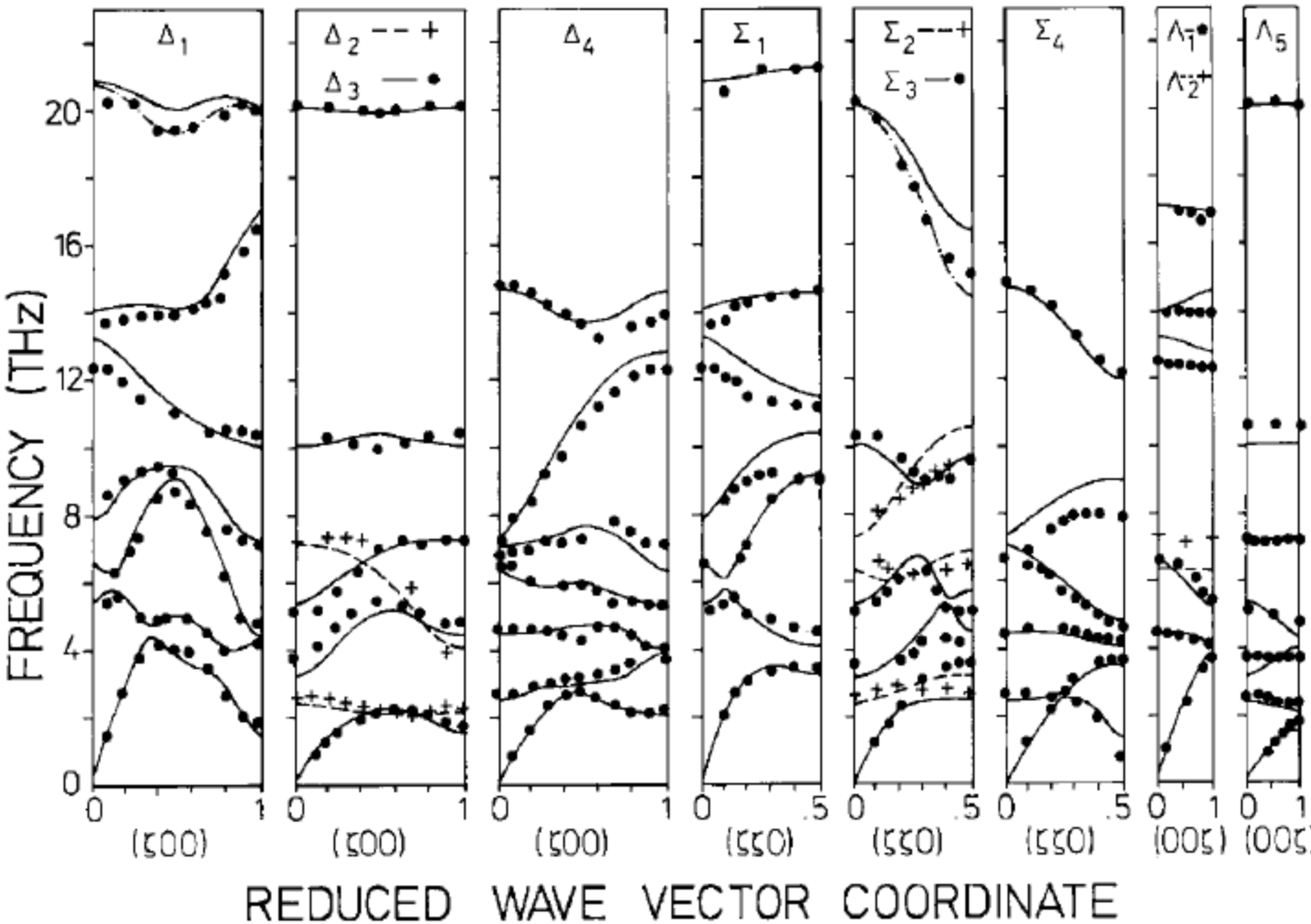
$$\mathcal{F}(\mathbf{Q}) = \sum_j \frac{\bar{b}_j}{\sqrt{m_j}}(\mathbf{Q}\cdot\boldsymbol{\xi}_{js})e^{i\mathbf{Q}\cdot\mathbf{d}_j}e^{-W_j}$$

$\boldsymbol{\xi}_{js}$ phonon eigenvector for atom *j* of mode *s*

m_j mass of atom *j*

$\mathbf{d}_j$ position of atom *j* in the unit cell

Phonon dispersions in La_2CuO_4



Pintschovius, phys. stat. sol. b (2005)

more on phonons

Atomic displacement of atom *j*:

$$\mathbf{u}_{js} = \frac{\boldsymbol{\xi}_{js}}{\sqrt{m_j}}$$

Eigenvector sum rule:

$$\sum_j |\boldsymbol{\xi}_{js}|^2 = 1$$

Acoustic mode at small $\mathbf{q}$:

$$\lim_{\mathbf{q}\to 0} \frac{\boldsymbol{\xi}_{ja}}{\sqrt{m_j}} = \frac{\hat{\mathbf{e}}}{M}$$

where $\hat{\mathbf{e}} = \mathbf{u}_{ja}/u_{ja}$ $\quad M = \sum_j m_j$

Finally:

$$\lim_{\mathbf{q}\to 0} \mathcal{F}^2_{\mathrm{acoustic}}(\mathbf{Q}) = \frac{G^2}{M} F_N^2(\mathbf{G})$$

Can be used to put cross section on absolute scale

Incoherent scattering

Elastic scattering:

$$\left.\frac{d\sigma}{d\Omega}\right|_{\mathrm{inc}} = \frac{N}{4\pi}\sum_j \sigma_{\mathrm{inc},j} e^{-2W_j}$$

Inelastic scattering from a Bravais lattice:

$$\left.\frac{d^2\sigma}{d\Omega_f dE_f}\right|_{\mathrm{inc}} = \frac{\sigma_{\mathrm{inc}}}{4\pi}\frac{k_f}{k_i}\frac{3N}{2m}\frac{e^{-2W}}{1-e^{-\hbar\omega/k_BT}}\frac{\langle(\mathbf{Q}\cdot\boldsymbol{\xi}_s)^2\rangle}{\omega}G(\omega)$$

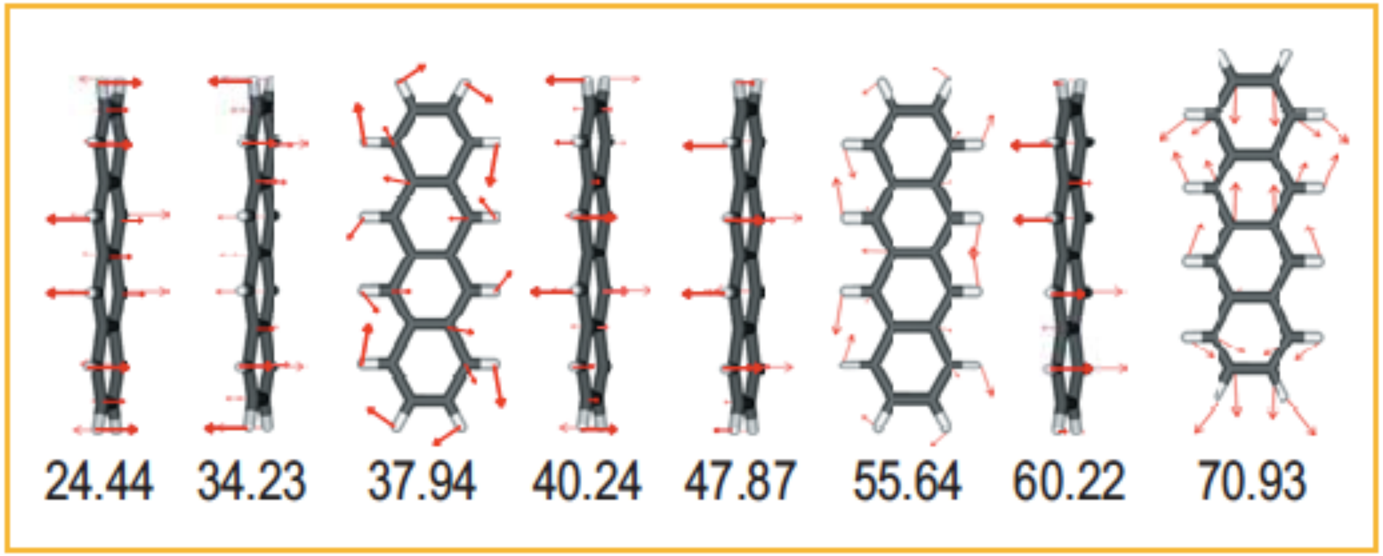
phonon density of states

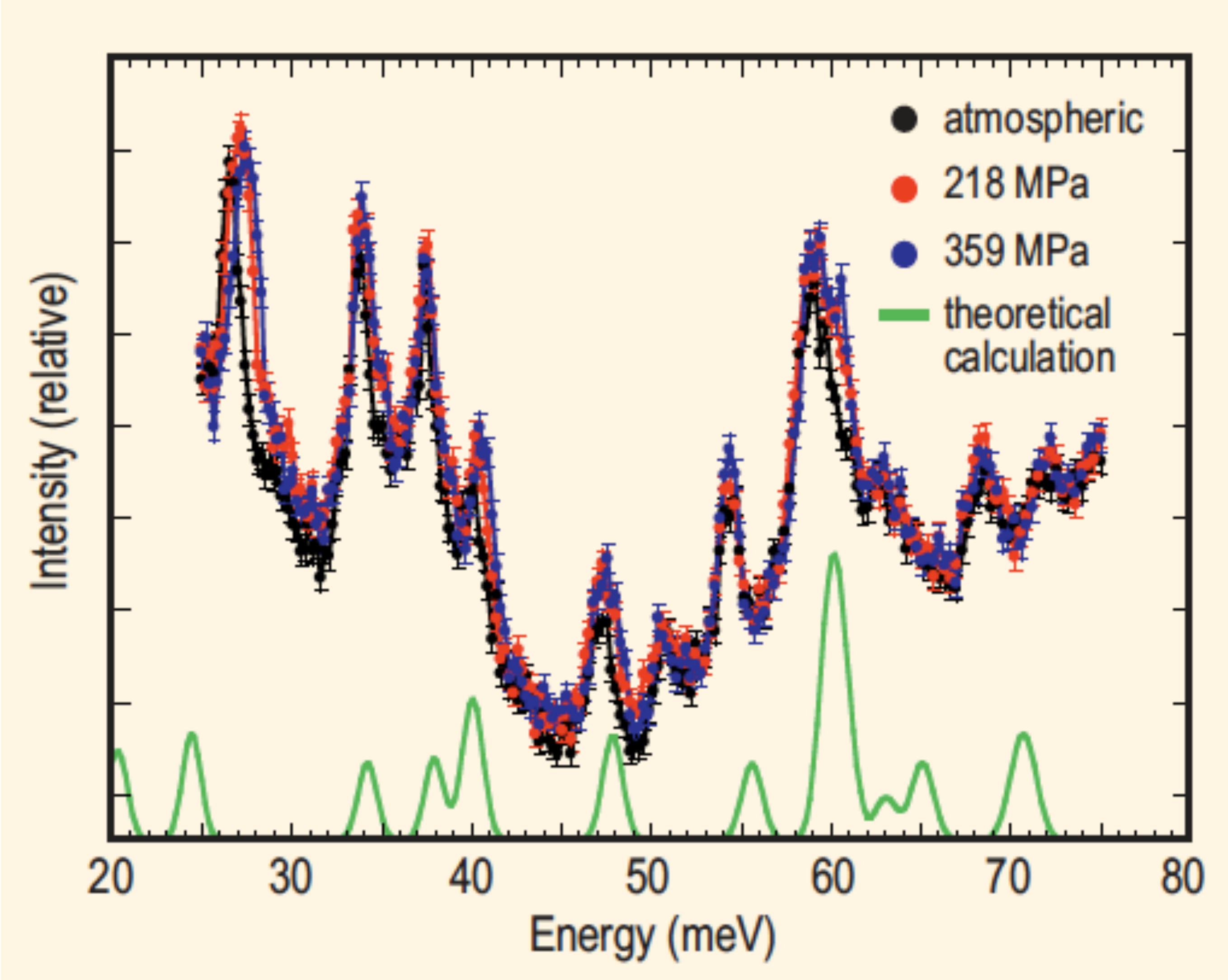
For a cubic crystal: $\langle(\mathbf{Q}\cdot\boldsymbol{\xi}_s)^2\rangle = \frac{1}{3}Q^2$

More generally:

$$\left.\frac{d^2\sigma}{d\Omega_f dE_f}\right|_{\mathrm{inc}} = \frac{k_f}{k_i}\sum_j \frac{\sigma_{\mathrm{inc},j}}{2m_j}\frac{e^{-2W}}{1-e^{-\hbar\omega/k_BT}}\sum_s \frac{|\mathbf{Q}\cdot\boldsymbol{\xi}_{js}|^2}{\omega_s}\delta(\omega-\omega_s)$$

Example: H modes in Tetracene





A.M. Pivovar *et al.*,
Chem. Phys. **325**, 138 (2006)

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