Neutron Scattering
with Examples from Cuprate Superconductors
Part I

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Quantum Science Summer School
Cornell University
June 21, 2018
> 30 years since discovery of high $T_c$ superconductivity

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$_{2-x}$La$_x$Cu$_2$O$_4$ (lower curve) and Ba$_{2-x}$La$_x$Cu$_2$O$_4$ (upper curve) 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:

$$\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$$

Zürich oxide
The High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory was built in the early 1960s because of the constant needs of scientists to always want "more". In the mid-1950s, the Brookhaven Graphite reactor was producing a number of important new results when that generation of scientists realized the need for a high flux reactor and started down the political, scientific, and engineering path that led to the HFBR.

- **1965 - HFBR first goes critical**
- **1999 - HFBR officially closed**
Crystal structure of La$_{2-x}$Ba$_x$CuO$_4$

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

“Lattice instability and high-$T_c$ superconductivity in La$_{2-x}$Ba$_x$CuO$_4$”

“Structural phase transformations and superconductivity in La$_{2-x}$Ba$_x$CuO$_4$”
Antiferromagnetism in La$_2$CuO$_4-\gamma$


Corporate Research Laboratories, Exxon Research and Engineering Company, Annandale, New Jersey 08801
(Received 4 May 1987)
The blind physicists and the superconductor

High $T_c$ Superconductivity

- Pairing
- Defect
- Ultrasonics
- STM
- Tunneling
- SQUID
- Photoemission
- Transport
- Applications
- Neutron Scattering

Model

THEORY

Properties of the Neutron

mass \quad m_n = 1.009 \text{ amu} \\
\approx m_p \approx 1838 \text{ } m_e

spin \quad 1/2

magnetic moment \quad 1.913 \mu_N \\
0.001 \mu_B

charge \quad 0
Table 2: Wavelength, frequency, velocity and energy relationships for neutrons.

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

\[ k = \frac{2\pi}{\lambda} \]

\[ E = \frac{\hbar^2 k^2}{2m_n} \]

\[ d\Omega_f = \text{differential of solid angle for } k_f \]

\[ dE_f = \text{energies between } E_f \text{ and } E_f + dE_f \]

\[ \frac{d^2\sigma}{d\Omega_f dE_f} = \text{probability of scattering into } d\Omega_f dE_f \]
Momentum and energy transfer

$$Q = k_f - k_i \quad \text{(momentum conservation)}$$

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

$$\hbar \omega = E_i - E_f \quad \text{(energy conservation)}$$

$$= \frac{\hbar^2}{2m_n} (k_i^2 - k_f^2)$$
Differential cross section

Fermi’s golden rule:

\[
\frac{d^2\sigma}{d\Omega_f dE_f} \bigg|_{\lambda_i \to \lambda_f} = \frac{k_f}{k_i} \left( \frac{m_n}{2\pi\hbar^2} \right)^2 |\langle k_f \lambda_f | V | k_i \lambda_i \rangle|^2 \delta(\hbar\omega + E_i - E_f)
\]

Born approximation (treat neutrons as plane waves):

\[
\langle k_f \lambda_f | V | k_i \lambda_i \rangle = V(Q) \langle \lambda_f | \sum_l e^{iQ \cdot r_l} | \lambda_i \rangle
\]

for a collection of atoms at positions \( r_l \)

\[
V(Q) = \int dr \ V(r) e^{iQ \cdot r}
\]

Nuclear scattering:

\[
V(Q) = \frac{2\pi\hbar^2}{m_n} b
\]
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:

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

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

Total scattering:

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

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

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

$$b_{inc} = \sqrt{\bar{b}^2 - \bar{b}^2}$$
Examples of coherent vs. incoherent

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Natural Abundance (%)</th>
<th>( b_{\text{coh}} ) (fm)</th>
<th>( b_{\text{inc}} ) (fm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H</td>
<td>100</td>
<td>( 7.79(1) )</td>
<td>( 0.12(3) )</td>
</tr>
<tr>
<td>(^1\text{H})</td>
<td>99.985</td>
<td>(-3.7390(11))</td>
<td>( 25.274(9) )</td>
</tr>
<tr>
<td>(^2\text{H})</td>
<td>0.015</td>
<td>( 6.671(4) )</td>
<td>( 4.04(3) )</td>
</tr>
<tr>
<td>(^3\text{H})</td>
<td>(12.32 a)</td>
<td>( 4.792(27) )</td>
<td>(-1.04(17) )</td>
</tr>
<tr>
<td>(^10\text{B})</td>
<td>20</td>
<td>( 5.30(4) - 0.213(2)i )</td>
<td>(-4.7(3) + 1.231(3)i )</td>
</tr>
<tr>
<td>(^{11}\text{B})</td>
<td>80</td>
<td>( 6.65(4) )</td>
<td>(-1.3(2) )</td>
</tr>
</tbody>
</table>
Scattering lengths in $\text{La}_{2-x}(\text{Ba},\text{Sr})_x\text{CuO}_4$

<table>
<thead>
<tr>
<th>Element</th>
<th>$b_{\text{coh}}$ (fm)</th>
<th>Z</th>
</tr>
</thead>
<tbody>
<tr>
<td>La</td>
<td>8.24</td>
<td>57</td>
</tr>
<tr>
<td>Ba</td>
<td>5.07</td>
<td>56</td>
</tr>
<tr>
<td>Sr</td>
<td>7.02</td>
<td>38</td>
</tr>
<tr>
<td>Cu</td>
<td>7.72</td>
<td>29</td>
</tr>
<tr>
<td>O</td>
<td>5.80</td>
<td>16</td>
</tr>
</tbody>
</table>

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:

\[
\frac{d^2\sigma}{d\Omega_f dE_f} = \frac{k_f}{k_i} \sum_{\lambda_i, \lambda_f} P(\lambda_i) \left| \langle \lambda_f | \sum_l e^{iQ \cdot r_l} | \lambda_i \rangle \right|^2 \delta(h\omega + E_i - E_f)
\]

\[P(\lambda_i) = \text{distribution of initial states}\]

Dynamical structure factor (Van Hove)

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

\[
S(Q, \omega) = \frac{1}{2\pi\hbar N} \sum_{ll'} \int_{-\infty}^{\infty} dt \left\langle e^{-iQ \cdot r_{l'}(0)} e^{iQ \cdot r_{l}(t)} \right\rangle e^{-i\omega t}
\]

\[
\langle \ldots \rangle = \sum_i P(\lambda_i) \ldots
\]
Coherent elastic nuclear scattering

Time-averaged structure factor:

\[
S_{\text{coh}}(Q, \omega) = \delta(\hbar \omega) \frac{1}{N} \left\langle \sum_{ll'} e^{iQ \cdot (r_l - r_{l'})} \right\rangle
\]

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

\[
S_{\text{coh}}(Q, \omega) = \delta(\hbar \omega) \frac{(2\pi)^3}{v_0} \sum_G \delta(Q - G)
\]

\(G = \text{reciprocal lattice wave vector}\)

\(v_0 = \text{volume per unit cell}\)

Coherent elastic cross section:

\[
\frac{d\sigma^{\text{el}}}{d\Omega}_{\text{coh}} = N \frac{(2\pi)^3}{v_0} \left( \frac{\bar{b}}{b} \right)^2 \sum_G \delta(Q - G)
\]
Generalizations

Impact of lattice vibrations:

\[ \langle e^{iQ \cdot r} \rangle = e^{-2W} \]

Debye-Waller factor

\[ W = \frac{1}{2} \langle (Q \cdot u)^2 \rangle \]

Average is over atomic displacements \( u \)

Multiple atoms per unit cell:

\[ \frac{d \sigma}{d \Omega} \bigg|_{el} = N \frac{(2\pi)^3}{v_0} \sum_G \delta(Q - G) |F_N(G)|^2 \]

Nuclear structure factor

\[ F_N(G) = \sum_j \tilde{b}_j e^{iG \cdot d_j} e^{-W_j} \]
Neutron Sources

- Neutrons live only in nuclei
  - Binding energy $\sim 10$ 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 $\sim e^{-E/kT}$
  - $kT \sim 20$ K $\sim 2$ meV cold neutrons
  - $kT \sim 300$ K $\sim 26$ meV thermal neutrons
  - 100 - 1000 meV epithermal neutrons
# Major Neutron User Facilities

<table>
<thead>
<tr>
<th>Reactors</th>
<th>Spallation sources</th>
</tr>
</thead>
<tbody>
<tr>
<td>HFIR, Oak Ridge, TN</td>
<td>SNS, Oak Ridge, TN</td>
</tr>
<tr>
<td>NCNR, Gaithersburg, MD</td>
<td>ISIS, Oxfordshire, UK</td>
</tr>
<tr>
<td>ILL, Grenoble, France</td>
<td>J-PARC, Tokai, Japan</td>
</tr>
<tr>
<td>FRM-II, Munich, Germany</td>
<td>SINQ, Villigen, Switzerland</td>
</tr>
<tr>
<td>OPAL, Lucas Heights, Australia</td>
<td>CSNS, Dongguan, China</td>
</tr>
<tr>
<td>JRR-3, Tokai, Japan (2020)</td>
<td>ESS, Lund, Sweden (2023)</td>
</tr>
</tbody>
</table>
**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:

\[ S(-Q, -\omega) = e^{-\hbar \omega / k_B T} S(Q, \omega) \]

Fluctuation-dissipation theorem gives:

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

Dynamic susceptibility:

\[ \chi(Q, \omega) = \chi'(Q, \omega) + i \chi''(Q, \omega) \]

\[ \chi''(Q, -\omega) = -\chi''(Q, \omega) \]

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

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

$$Q = G + q$$

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

Acoustic modes at small $q$

$$\omega_{qs} \approx v_s q$$

Phonon dispersions are the same in every Brillouin zone but the intensities can vary with $Q$
Phonon susceptibility

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

\[
F(Q) = \sum_j \frac{\bar{b}_j}{\sqrt{m_j}} (Q \cdot \xi_{js}) e^{iQ \cdot \mathbf{d}_j} e^{-W_j}
\]

- $\xi_{js}$: phonon eigenvector for atom $j$ of mode $s$
- $m_j$: mass of atom $j$
- $d_j$: position of atom $j$ in the unit cell
Phonon dispersions in $\text{La}_2\text{CuO}_4$

more on phonons

Atomic displacement of atom $j$:

$$u_{js} = \frac{\xi_{js}}{\sqrt{m_j}}$$

Eigenvector sum rule:

$$\sum_j |\xi_{js}|^2 = 1$$

Acoustic mode at small $q$:

$$\lim_{q \to 0} \frac{\xi_{ja}}{\sqrt{m_j}} = \hat{e} = \frac{\hat{e}}{M}$$

where

$$\hat{e} = u_{ja}/u_{ja}$$

$$M = \sum_j m_j$$

Finally:

$$\lim_{q \to 0} F_{\text{acoustic}}^2(Q) = \frac{G^2}{M} F_N^2(G)$$

Can be used to put cross section on absolute scale
Incoherent scattering

Elastic scattering:

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

Inelastic scattering from a Bravais lattice:

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

For a cubic crystal:

\[
\left\langle (Q \cdot \xi_s)^2 \right\rangle = \frac{1}{3} Q^2
\]

More generally:

\[
\left. \frac{d^2\sigma}{d\Omega_f dE_f} \right|_{\text{inc}} = \frac{k_f}{k_i} \sum_j \frac{\sigma_{\text{inc},j}}{2m_j} \frac{e^{-2W}}{1 - e^{-\hbar\omega/k_BT}} \sum_s \frac{|Q \cdot \xi_{js}|^2}{\omega_s} \delta(\omega - \omega_s)
\]
Example: H modes in Tetracene

A.M. Pivovar et al.,
End of Part 1