Neutrons and High Pressure

John B. Parise
Stony Brook University, NY
RANGE OF PRESSURES IN THE UNIVERSE

- Hydrogen gas in intergalactic space: $10^{-32}$ atm
- Interplanetary space: $10^{-2}$ atm
- Atmosphere at 300 miles: $10^{-8}$ atm
- Center of Jupiter: $10^8$ atm
- Center of Sun: $10^{16}$ atm
- Center of white dwarf: $10^2$ atm
- Center of neutron star: $10^3$ atm
- Deepest ocean: $10^4$ atm
- Water vapor at triple point: $10^6$ atm
- Atmospheric pressure (sea level): $10^8$ atm
- Center of the Earth: $10^{10}$ atm
- Best mechanical pump vacuum: $10^{-8}$ atm
Why Pressure? Science Issues Addressed

- Nature of dense hydrogen - *cryogenic to brown dwarf conditions*
- Composition, elasticity, and thermal state of Earth’s core
- Structures of complex hydrous phases
  - *Clathrates, molecular compounds, hydrous silicates, metal hydrides*
- **Hydrogen bonding** - *Organic & inorganic systems, inc. liquids*
- Supercritical fluids and liquids
  - *Structure and dynamics and effect on chemical reactions*
- Structure and dynamics of *silicate melts and glasses*
- Planetary ices - *Structure, strength, and dynamics*
- Influence of pressure and stress on *magnetic properties*
- Structure and dynamics of *nanomaterials under pressure*
- General *phase transition studies*
- Chemical kinetics and reaction mechanism - *inner vs. outer sphere*
- Your favorite application here
Early cells (courtesy JD Jorgensen)


100 kbar opposed-anvil press for time-of-flight neutron powder diffraction

Al₂O₃ anvils with 1 inch diameter faces

Sample volume = 0.05 cm³

(0.6 cm diam. x 0.3 cm thick)
From early experiments at reactors

- Jim recognized early
  - Clear interference from cell a huge problem in angle dispersive mode
- Solutions
  - fixed exit cells (spallation or ED rather than AD scattering)
  - either live with the interference or rethink alternatives
- Other considerations
  - Gas cells (low P but high precision)
  - Solid media cells (high P) but also high noise
Early fixed angle time of flight set up

High-Pressure TOF Powder Diffractometer at Argonne's CP-5 Research Reactor
Gas Pressure Cell for Time-of-Flight Neutron Diffraction

Gadolinium-epoxy shielding

SECTION "A-A"

SECTION "B-B"

10 CM.

(Includes work done at the MTR Research Reactor at Argonne West in the late 1960's and later work at Argonne National Laboratory's CP-5 Reactor and Intense Pulsed Neutron Source)

- Pressure Dependence of the Morin Transition in α-Fe2O3 to 26 kbar, T. G. Worlton, R. B. Bennion, and R. M. Brugger, Phys. Lett. 24A, 653 (1967)
- "I believe this is the first Rietveld refinement done in the US. The authors were not aware of Rietveld’s work, so they did not cite his paper. They developed a least-squares fitting routine on their own and used it to compare different structural models to the data."
- 1977
1978
**Perhaps the most highly cited paper from the early days.
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**The first high-pressure study of an electronic topological (Lifshitz) transition.
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**Among the first in situ neutron diffraction work on the high-pressure phases of ice.
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1975
Structure and Order Parameters in the Pressure Induced Continuous Transition in TeO2, T. G. Worlton, R. A. Beyerlein, Phys. Rev. B 12, 1899-1907 (1975)

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**At the time, this was a pretty complex structure to determine from high-pressure data.

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Pressure-Induced Structural Changes in La1.85Sr0.15CuO4, S. Pei, J. D. Jorgensen, D. G. Hinks, B. D. Dabrowski, P. Lightfoot, and D. R. Richards, Physica C 169, 179-183 (1990)


1990


**A classic study of pressure effects in cuprate superconductors and also the first description of the helium gas pressure cell that we now use.

Pressure-Induced Structural Changes in La1.85Sr0.15CuO4, S. Pei, J. D. Jorgensen, D. G. Hinks, B. D. Dabrowski, P. Lightfoot, and D. R. Richards, Physica C 169, 179-183 (1990)


**A very nice short review if you can get your hands on the conference proceedings.

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Pressure-Induced Structural Changes in Superconducting \( \text{HgBa}_2\text{Ca}_1-\text{Cu}_2\text{O}_{n+2+\delta} \) (n=1,2,3) Compounds, B. A. Hunter, J. D. Jorgensen, J. L. Wagner, P. G. Radaelli, D. G. Hinks, H. Shaked, R. L. Hitterman, and R. B. Von Dreеле, Physica C 221, 1-10 (1994)

**A key paper for learning how pressure can increase \( T_c \) in a cuprate superconductor.

Pressure-Induced Phase Transition and Pressure Dependence of Crystal Structure in Low (a) and Ca/Al-doped Cristobalite, J. B. Parise, A. Yeganeh-Haeri, D. J. Weidner, J. D. Jorgensen, and M. A. Saltzberg, J. Appl. Phys. 75, 1361-1367 (1994)


Structural Effects of Hydrostatic Pressure in \( \text{Sr}_1\text{x}_\text{Mx}\text{CuO}_2 \) (M=La,Ca) and \( \text{Sr}_4\text{Cu}_6\text{O}_{10} \), H. Shaked, Y. Shimakawa, B. A. Hunter, P. G. Radaelli, B. Dabrowski, R. L. Hitterman, J. D. Jorgensen, P. Han, D. A. Payne, S. Kikkawa, G. Er, F. Kanamaru, Phys. Rev. B, 50, 12752-12759, (1994)

1995

**A very interesting paper about how rare gas intercalation affects the compressibility of C60.


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**A nice summary of how high-pressure work on cuprate superconductors leads to an understanding of how \( T_c \) is optimized in these materials.


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Compressibility, Phase Transitions, and Oxygen Migration in the Zirconium Tungstate, \( \text{ZrW}_2\text{O}_8 \), J. S. O. Evans, Z. Hu, J. D. Jorgensen, D. N. Argyriou, S. Short, A. W. Sleight, Science 275, 61-65 (3 January 1997)

**I think this is the first high-pressure diffraction paper on this classic negative thermal expansion material.

Rare-Gas Intercalation into Fullerene Interstices, G. H. Kwei, J. D. Jorgensen, B. Morosin, Fullerene Science and Technology 5, 243-256 (1997)

Argonne High Pressure Neutron Diffraction Publications:
(1966-2006, compiled by J. D. Jorgensen)

1994 - Pressure-Induced Structural Changes in Superconducting HgBa$_2$Ca$_{n-1}$Cu$_n$O$_{2n+2+2\alpha}$ (n=1,2,3) Compounds, B. A. Hunter, J. D. Jorgensen, J. L. Wagner, P. G. Radaelli, D. G. Hinks, H. Shaked, R. L. Hitterman, and R. B. Von Dreele, Physica C 221, 1-10 (1994) **A key paper for learning how pressure can increase $T_c$ in a cuprate superconductor.


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Thermal Expansion and Compressibility in Superconducting Na$_x$CoO$_2$•4xO$_2$ (x≈1/3): Evidence for Pressure-Induced Charge Redistribution, J. D. Jorgensen, M. Avdeev, D. G. Hinks, P. W. Barnes, and S. Short, Phys. Rev. B 72, 224515 (21 December 2005) **A nice example of how pressure can drive dramatic changes in hydrogen bonding resulting in charge transfer and modification of transport and superconducting properties.

2006 - Pressure-induced ferroelectric to antiferroelectric phase transition of PZT95/5(2Nb): A neutron powder diffraction and dielectric study, Maxim Avdeev, James D. Jorgensen, Simine Short, George A. Samara, Eugene L. Venturini, Pin Yang, and Bruno Morosin, Phys. Rev. B. 73, 064105 (9 February 2006) **Upon going through a pressure-induced phase transition, a mixed phase is seen in which the ferroelectric phase has a substantially enhanced polarization due to grain-interaction stresses.
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<td>Room Temperature Compressibility of C$_{60}$: Intercalation Effects with He, Ne and Ar</td>
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<td>Structural Parameters for the Suspected Pressure-Induced Electron Transition in InBi</td>
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Order Parameter and Critical Exponent for the Pressure-induced Phase Transitions in \( \text{ReO}_3 \), J.-E. Jørgensen, J. D. Jorgensen, B. Batlogg, J. P. Remeika, and J. D. Axe, Phys. Rev. B 33, 4793-4798 (1986) **A truly beautiful piece of work on the pressure-induced soft-mode transition in \( \text{ReO}_3 \). This is one of the few cubic perovskites that does what perovskites are predicted to do at high pressure.
J. D. Jorgensen’s High Pressure legacy

- There are important insights provided by studying materials @ HP
- Good signal-to-noise is essential
- How to get it is the challenge
- Jim’s solutions
  - spallation
  - quiet gas pressure cells
- Unstinting support for instrument development (including SNAP at SNS)
J. D. Jorgensen’s High Pressure legacy
The HP community strives to match what Jim did - at higher pressures in solid media devices (contributors to this talk in parentheses)
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Increase sample volume - PE initiative (thanks to Besson/Nelmes/Klotz)
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Focusing to improve signal (Gene Ice)
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- Single crystal studies in gem cells? (Goncharenko, Xu, Mao)
  - area detectors and white beams (ILL, McIntyre)
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- Bringing it all together - the SNAP beamline (Tulk)
HP Devices: the DIA and PE cells
Pressure generation

- \( P = \frac{F}{A} \)
  - Diamond anvils - small volume high \( P \)
  - Paris-Edinburgh cell
  - Large - volume (80 mm\(^3\) +) at 10+ GPa
Pressure generation

Diamond anvils - small volume high P
Paris-Edinburgh cell
Large - volume (80 mm$^3$) at 10+ GPa

HP Devices: the DIA and PE cells

Be backing plate
X-rays

Gasket (W, Re, steel)
Sample

Be backing plate
X-rays

Secondary scattering from Be backing plate
Diamond reflection

(a) (b) (c) (d)

MSA Neutrons, Dec 7
Pressure generation

Diamond anvils - small volume high P

Paris-Edinburgh cell

Large volume (80 mm$^3$ +) at 10+ GPa

HP Devices: the DIA and PE cells.

Be backing plate

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INCIDENT NEUTRONS

TO 90° DETECTOR BANKS

(a)
Pressure generation

- Diamond anvils - small volume high P
- Paris-Edinburgh cell
- Large volume (80 mm$^3$) at 10+ GPa

HP Devices: the DIA and PE cells.

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sample

INCIDENT NEUTRONS

TO 90° DETECTOR BANKS

3 cm

(a)

(a)
Pressure generation

P = F/A

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Paris-Edinburgh cell

Large - volume (80 mm$^3$) at 10+ GPa

HP Devices: the DIA and PE cells

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Sample

(a)

INCLUDENT

NEUTRONS

3 cm

TO 90° DETECTOR BANKS

15 cm

Be backing plate

X-rays
The PE cell first (V3) and later (V13) generations (Klotz, Hamel)
BEYOND THE STATE OF THE ART: 300 - 500 GPa? NEW WINDOWS ON PLANETARY MATERIALS (Xu, Mao, Hemley) - GPL

- New ‘Transparent’ Gaskets
- Direct Measure of Stress-Strain
- New High-Pressure Probes
- Transport Measurements
Four new phases discovered up to 6 GPa. Titan models assume negligible compression and no phase transitions.

Phase VI is a simple bcc structure with substitutional site disorder of water and ammonia.

Methane hydrate
Central to models of the origin of Titan’s atmosphere

Previously thought to decompose into ice and methane in the 1-2 GPa range

- Two new high pressure hydrates
  - phase II (H₂O)₃.₅(CH₄)
  - phase III (H₂O)₂(CH₄)

stable to at least 10 GPa  Collaboration between University of Edinburgh and NRC Ottawa
Next generation - HP at low $T(< 20 \text{ K})$
Next generation - HP at low T(< 20 K)

This is clearly NOT a DOE facility
Properties - magnetic moment

Applications

**Ferrimagnetic magnetite**
\[ \text{Fe}_3\text{O}_4 \]

**Antiferromagnetic manganese oxide**
\[ \text{MnO} \]
Properties - magnetic moment

Applications

Determine magnetic structure from powder and single crystal patterns

Ferrimagnetic magnetite
\[ \text{Fe}_3\text{O}_4 \]

Antiferromagnetic manganese oxide
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Properties - magnetic moment

Applications

- Determine **magnetic structure** from powder and single crystal patterns
- Combination of polarized neutrons + single crystal powerful for complex (incommensurate, helical systems)

**Ferrimagnetic magnetite** $\text{Fe}_3\text{O}_4$

**Antiferromagnetic manganese oxide** $\text{MnO}$
Properties - magnetic moment

Applications

- Determine magnetic structure from powder and single crystal patterns
- Combination of polarized neutrons + single crystal powerful for complex (incommensurate, helical systems)
- Magnetism diagnostic for (nuclear) psuedo-symmetric phase transitions.

![Ferrimagnetic magnetite Fe₃O₄](image1)

![Antiferromagnetic manganese oxide MnO](image2)
FeS phase diagram

Fei, Prewitt, Mao, and Bertka (1995), Science
FeS phase diagram

Magnetic Scattering for FeS - a lucky break
Marshall et al, ISIS
Magnetic Scattering for FeS - a lucky break
Marshall et al, ISIS

This is subtle! and most time, with powders, we only know the moment is either in the plane or not. Best to use single crystals
Neutron mass ~ proton mass: shift spectrum by allowing neutrons to come to thermal equilibrium with proton-rich (CH$_4$, H$_2$, H$_2$O, D$_2$O) condensed matter at different temperatures)

(a) Reactor Neutrons

(a) Spallation Neutrons
Neutron mass ~ proton mass: shift spectrum by allowing neutrons to come to thermal equilibrium with proton-rich (CH$_4$, H$_2$, H$_2$O, D$_2$O) condensed matter at different temperatures)

(a) Reactor Neutrons

- 20 K
- 300 K
- 2000 K

Typical monochromator cut

Optimized to low E (high d-space) magnetism, phonons, INS

(a) Spallation Neutrons

- 20 K
- 290 K

Relative Flux of Neutrons vs. Wavelength ($\lambda$, Å) and Energy (E, meV)
The PE cell at reactor sources - advantages

Neutron mass $\sim$ proton mass: shift spectrum by allowing neutrons to come to thermal equilibrium with proton-rich ($\text{CH}_4$, $\text{H}_2$, $\text{H}_2\text{O}$, $\text{D}_2\text{O}$) condensed matter at different temperatures

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- 20 K
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Wavelength ($\lambda$, Å)

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magnetism, phonons, INS

Optimized high $E$ (low $d$-space)
high precision structural studies (PDF for eg)
The PE cell at reactor sources - advantages

**Neutron mass ~ proton mass: shift spectrum by allowing neutrons to come to thermal equilibrium with proton-rich (CH$_4$, H$_2$, H$_2$O, D$_2$O) condensed matter at different temperatures)**

(a) Reactor Neutrons

- 20 K
- 300 K
- 2000 K

(b) Spallation Neutrons

- 20 K
- 290 K

Optimized to low E (high d-space) magnetism, phonons, INS

Optimized high E (low d-space) high precision structural studies (PDF for eg)

These are generalizations
The PE cell at reactor sources - advantages
Figure 1: Pressure dependence of the CoO diffraction patterns measured at 300 K on the D20 diffractometer. The appearance and increase of the magnetic contribution is well evidenced with the \((1/2,1/2,1/2)\) magnetic peak intensity variation. The measurement time for each pattern is 40 min. The data have been corrected for the nuclear peak.

Figure 2: Low angular range zoom of the CoO diffraction pattern measured at 300 K under pressure. The magnetic contributions are marked with arrows.

Figure 3: Pressure dependence of the CoO magnetization at 300 K, derived from Rietveld fits to the patterns shown in figure 2. The line is a guide for the eye.
Other ambiguous magnetic structure

Above $T_M$

$T > 250$ K; low $P$

Other ambiguous magnetic structure

Above $T_M$

$T > 250 \text{ K}; \text{ low } P$

Morin transition

Low $T$ (High $P$?)

Other ambiguous magnetic structure

Above $T_M$

$T > 250 \text{ K; low } P$

Morin transition

Low $T$ (High $P$?)

Pressure (GPa)

Angle to $C_{\text{hexagonal}}$ (°)

Arbitrary Intensity


nano-hematite (this study)

Goncharenko et al. 1995

$2\theta(\text{°})$
Other ambiguous magnetic structure

Above $T_M$

$T > 250$ K; low P

Morin transition
Low T (High P?)

Other ambiguous magnetic structure

Above $T_M$

$T > 250 \, K; \, low \, P$

Morin transition

Low $T$ (High $P$)

Ambiguity over where in basal plane moment is pointing - need single crystal

Science opportunities: Kondo-lattice system CeSb: Rossat-Mignod and coworkers at ILL, Grenoble

Most complex magnetic phase diagram known so far:
consists of sixteen phases.
At H = 0 the low temperature Phase is the type-IA phase.
The rest are modulated AFP phases containing paramagnetic planes.

Coworkers: P. Burlet, J. Rossat-Mignod, C. Vettier, in ILL clamp cell
Important results:
Stabilization of type I AF phase.
Disappearance of AFP phases.
Type I and type IA at high pressure.
CeSb behaves like CeBi at high P, containing paramagnetic planes.

High P studies to 2 GPa

Graphical representation of the magnetic phase diagram:
- AFF1, AFF2, AFP, AF, P
- Magnetic field (KoE) vs. Temperature (K)
- Pressure (kbar) vs. Temperature (K)
- Neutron and magnetization symbols

MSA Neutrons, Dec 7
Science opportunities: Kondo-lattice system CeSb: Rossat-Mignod and coworkers at ILL, Grenoble

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- high $P$ studies to 2 GPa
- Coworkers: P. Burlet, J. Rossat-Mignod, C. Vettier, in ILL clamp cell

Important results:
- Stabilization of type I AF phase.
- Disappearance of AFP phases.
- Type I and type IA at high pressure.
- CeSb behaves like CeBi at high $P$, containing paramagnetic planes.

Complex structures need resolution by SCND; also to see diffuse scattering.
FeTaO$_6$, Chung, Balakrishnan, Visser & Paul (Warwick), McIntyre (ILL)
3-D antiferromagnetic order at 8 K, 2-D order above 8 K (ALL room pressure).
Difficult to imagine interpretation of complex magnetic structure from powders

A: 2 K with predicted nuclear pattern superimposed

B: 10 K - a hint of diffuse scattering at low Q
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A: 2 K with predicted nuclear pattern superimposed
B: 10 K - a hint of diffuse scattering at low Q
C: 10 K minus 2 K -> rods of magnetic scattering
High pressure single crystal neutron scattering: The way forward for complex hydrous minerals and magnetism

- Smaller samples
- Brighter beams
  - More flux (SNS)
  - Smaller beams (focusing)
- More sensitive, larger area, lower noise, detectors
- Laue technique
The Spallation Neutron Source (SNS) construction project will conclude in 2006. At 1.4 MW it will be the world’s most powerful source of neutrons and the world’s leading facility for neutron scattering. For what science will it be used?
(2) Greater brightness: Neutron Focusing

1 x 3 mm BN slit ~ 4 m from target
Testing and Optimization of neutron mirrors

- A new prototype microfocusing system tested.
- Measured spot size (90 x 90 um) with roughly 30 mrad on sample.
- Diffraction data from free standing 300x300 μm samples and from 200 um FeO single crystal sample in a panoramic cell--under pressure

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With KB mirrors in place

90 x 90 micron beam

SCATTERING VOLUME $\sim 0.0045 \text{ mm}^3$
Forserite - Mg$_2$SiO$_4$
$\Phi = 170^\circ$

Without KB mirrors in place

Beam much larger than sample

SCATTERING VOLUME $\sim 0.05 \text{ mm}^3$
Forserite - Mg$_2$SiO$_4$
$\Phi = 170^\circ$
Mirrors + cell

Figure 2 Panoramic cell
Mirrors + cell

Figure 2 Panoramic cell
J. D. Jorgensen’s High Pressure legacy

- Focusing (Gene Ice ORNL)

- From this
J. D. Jorgensen’s High Pressure legacy

- Focusing (Gene Ice ORNL)

- From this – to this
The way forward

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- Brighter beams
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- More sensitive, larger area, lower noise, detectors
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High pressure cells on Vivaldi  (white beam instrument, curved IP detector)

Area Detectors and white beams (VIVALDI, McIntyre ILL)

Closed Instrument

Mounting pressure cell in instrument

Note the conical binding rings
Laue diffraction pattern from a 0.5 mm$^3$ natrolite sample in a moissanite-anvil cell: a) $\varphi = 65^\circ$, exposure time 1 hr

One picture says it all (McIntyre et al, 2005)
CCDs at neutron (reactor) source

- First test last week
Bringing it all together - Spallation Neutrons And Pressure (SNAP) Instrument Components - synchrotron like

Sample Positioning Assembly

Detector Banks

PE Cell w/ Sample

Sample Positioning Assembly

Mirrors
Bringing it all together - Spallation Neutrons And Pressure (SNAP) Instrument Components - synchrotron like

Sample Positioning Assembly

Detector Banks

PE Cell w/ Sample

Sample Positioning Assembly

Mirrors
SNAP on schedule to take beam in 2008
Bright future for HP neutrons
Bright future for HP neutrons

Unique properties of neutron plus:

- Potentially revolutionary developments in
  - Sources - ISIS 2nd target, SNS, ILL upgrade
  - Detectors - esp ILL
  - Focusing - ORNL
  - HP cells - PE group, Saclay
Bright future for HP neutrons

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↓

A bright future especially in complementary studies of
- Structure
  - light elements
  - Isotope derived partials (glasses and melts)
  - Magnetic materials
- Those momentum transfers not accessible by inelastic X-ray scattering
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MSA Neutrons, Dec 7
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