Science using BESL at the APS

WORKSHOP ON SCIENCE WITH HIGH ENERGY X-RAYS

August 10, 2004

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Supported by NASA, NSF and DOE

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High Temperature Diffraction Studies

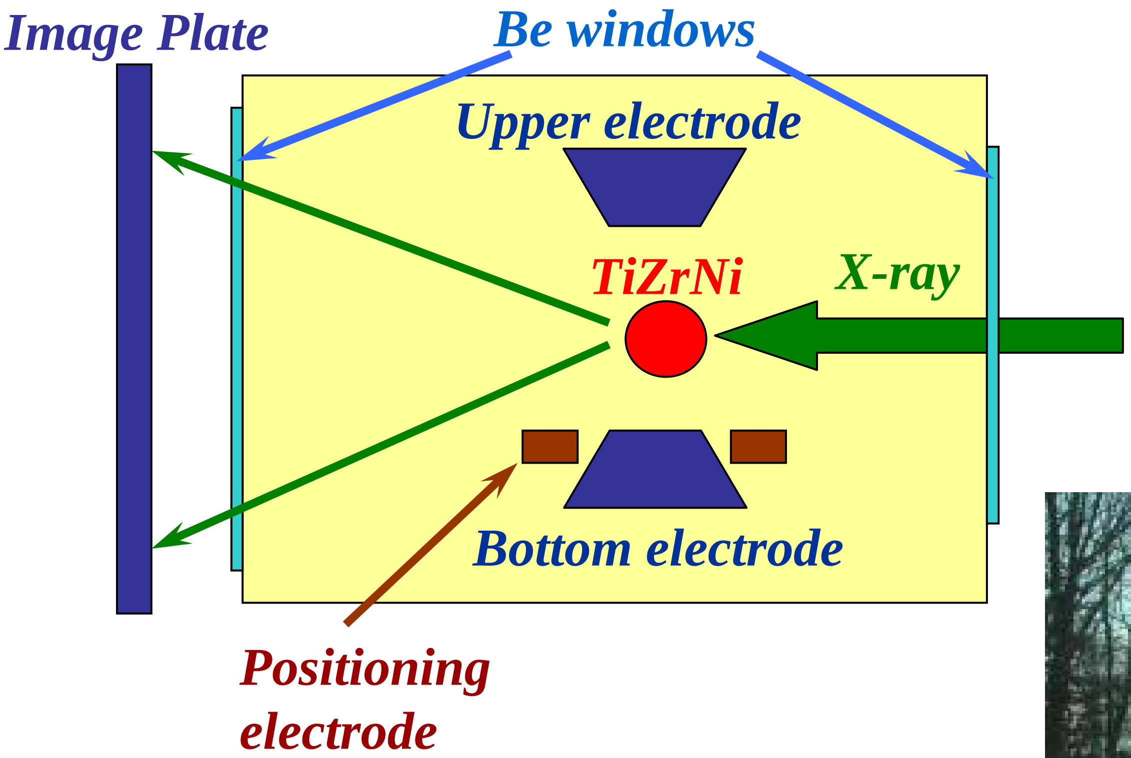
- **Structures of high temperature crystal phases**
- **Phase diagrams**
- **Phase formation sequence**
- **Transient metastable phase formation**
- **Liquid structures**
- **….**

Container and atmosphere contamination complicate interpretation

Containerless Processing {

Aerodynamic Levitation
Electromagnetic Levitation
Electrostatic Levitation

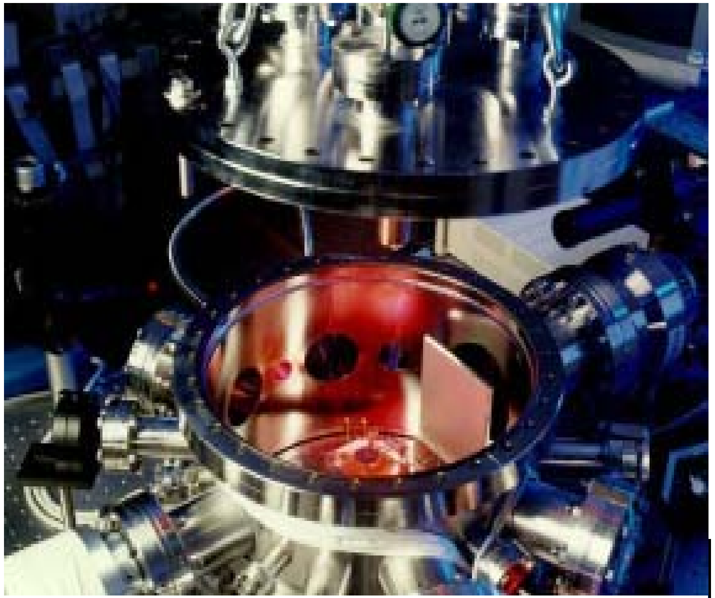
BESL
(Beamline Electrostatic Levitation)



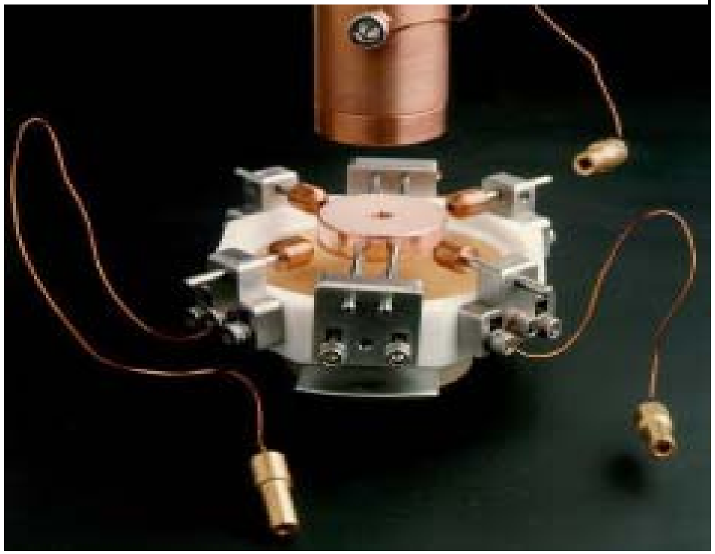
Wash. U., NASA, Ames Lab

K. F. Kelton, A. K. Gangopadhyay, G.W. Lee, J.R. Rogers, M. B. Robinson, R. W. Hyers, T. J. Rathz, A.. I. Goldman, D. S. Robinson

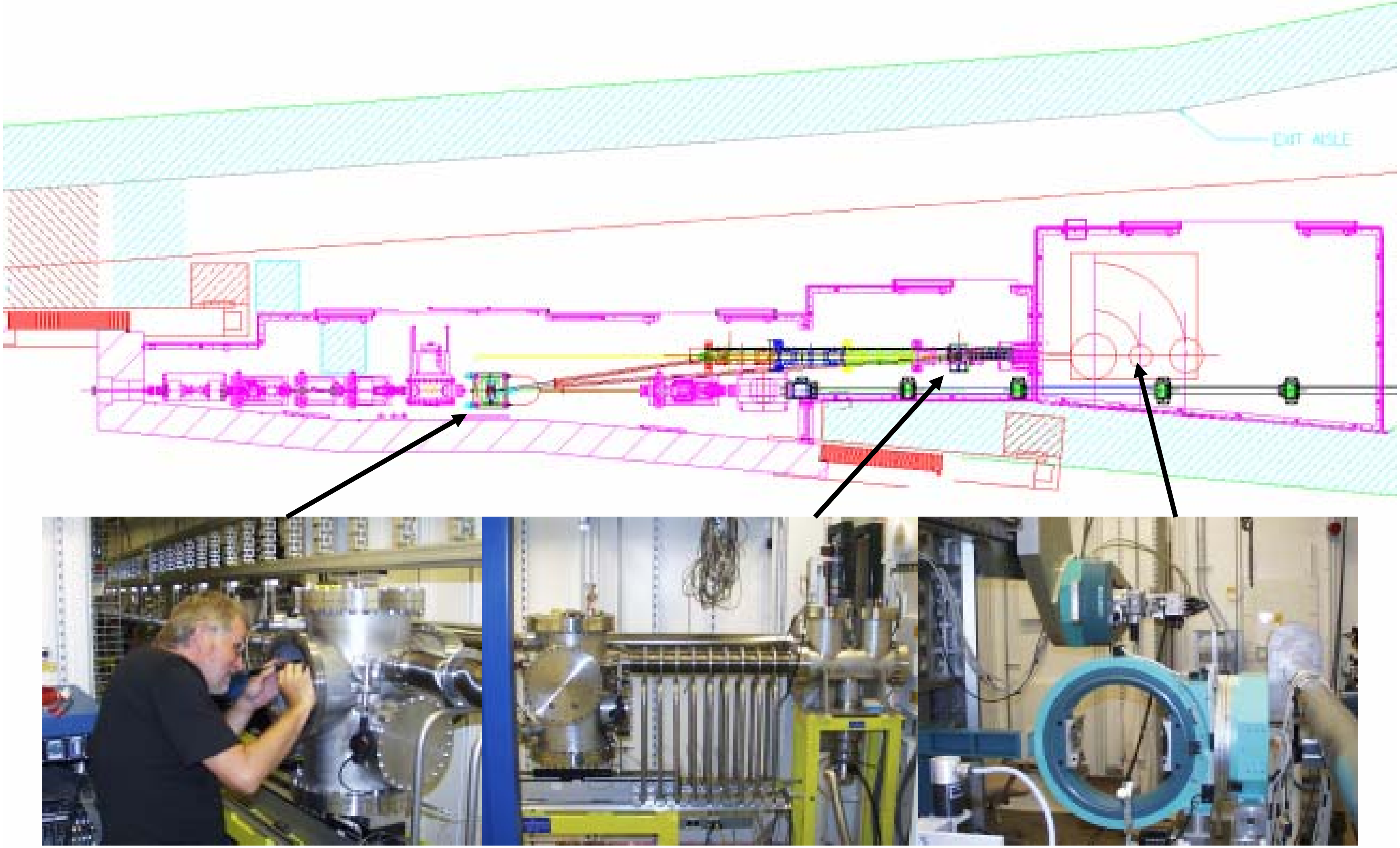
Electrostatic levitation (ESL)
- NASA Marshall Space Flight Center -



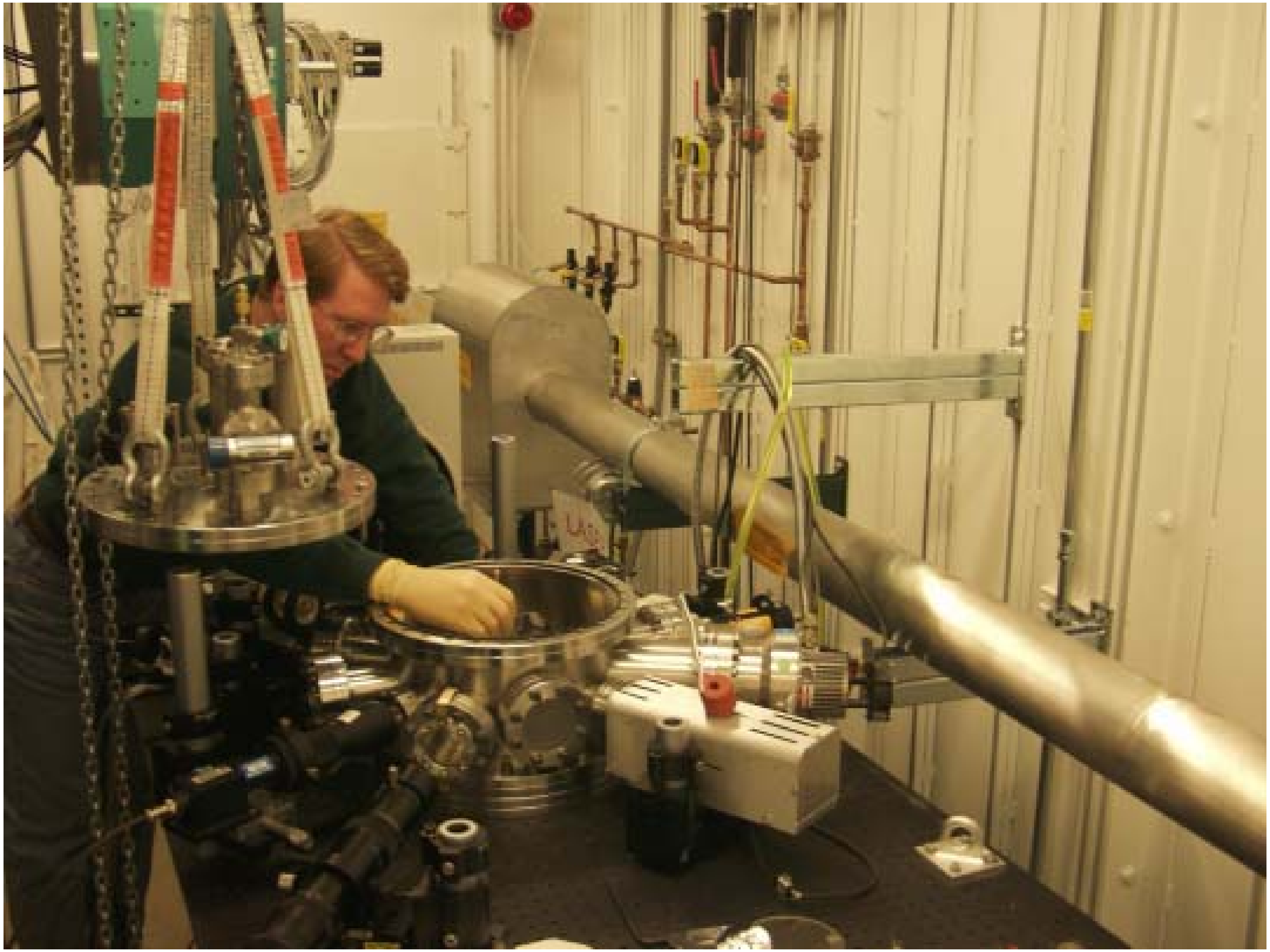
- Sample levitated in high vacuum ($\approx 10^{-8}$ torr)
- Add surface charge on sample by induction
- Maintain surface charge with ultraviolet lamp
- Apply large dc-field to generate sufficient force to counter gravity
- Fast feed-back mechanism to stabilize sample position (three independent sets of electrodes for x, y, and z positioning)
- Lasers used to heat the sample



6ID-D High Energy Side Station



BESL in 6ID-D at MUCAT



Detectors

GE Angio Detector

$\geq$30 ms collection times

CsI scintillator with amorphous Si on Glass

Based a medical technology

40 cm x 40 cm detection area

MAR 345

$\geq$ 1 s collection times

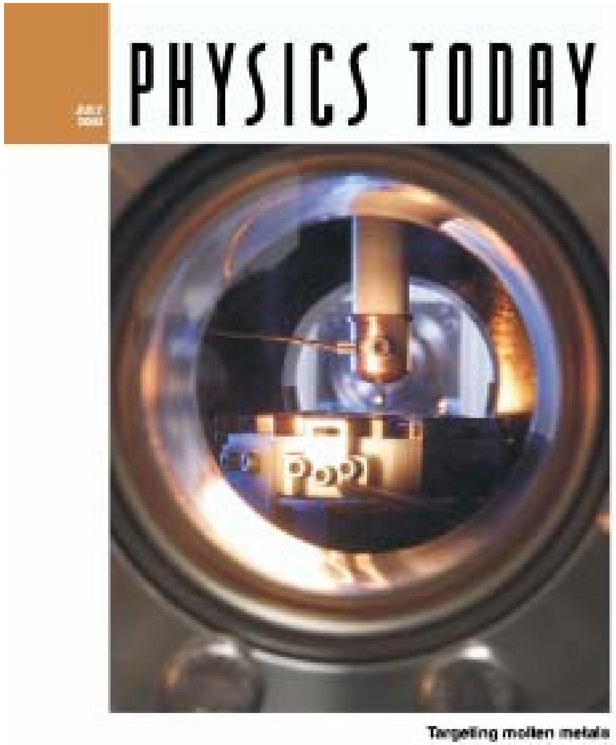
Phosphor image plate technology

30 second read times

34.5 cm x 34.5 cm detection area

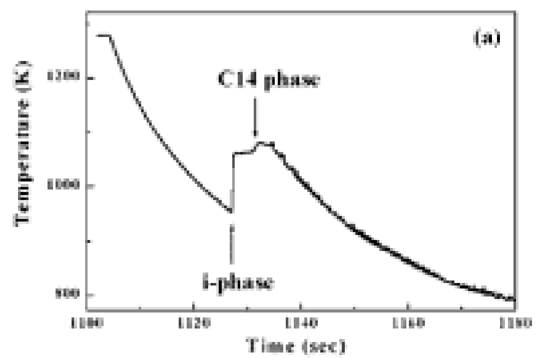
Experiments at the APS Vindicate a 50-Year-Old Explanation of How Liquid Metals Resist Solidification

K. F. Kelton, et al., Phys. Rev. Lett., ***90****, 195504/1-4 (2003)*

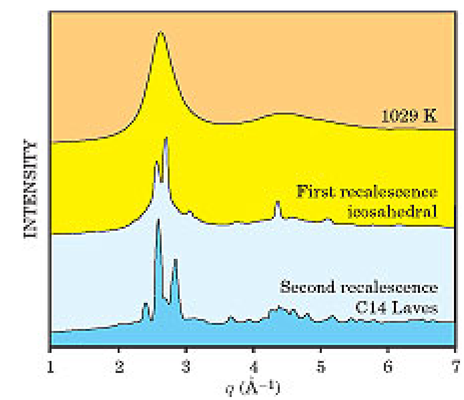


In-situ synchrotron x-ray diffraction studies of droplets of a TiZrNi liquid alloy that were levitated and heated in high vacuum demonstrated that icosahedral short-range atomic order exists and becomes more developed with decreasing temperature below the melting temperature. The increased icosahedral order favors the transformation of the liquid to a metastable icosahedral quasicrystal phase, demonstrating a clear connection between the nucleation barrier and the local structure of the liquid, and verifying Frank's hypothesis in this alloy. (Work done in the MUCAT Sector at the APS)

Many are familiar with the observation, first made by Fahrenheit, that liquid water can be cooled by a significant amount below its freezing temperature before ice forms. In 1952, David Turnbull showed that this ability to undercool was a common property, occurring even in metallic liquids. To explain this, Charles Frank proposed that the atoms in a liquid metal arrange themselves with the local symmetry of an icosahedron, a Platonic solid consisting of 20 tetrahedra (4-sided pyramid shaped polyhedra) arranged around a common center.



Cooling curves for showing two recalescence events (indicated by arrows). The first event corresponds to the nucleation and rapid growth of a metastable icosahedral quasicrystal; the second event indicates the transformation to the stable C14 crystal phase.



Diffraction patterns taken at the first and second recalescence events depicted in the previous figure

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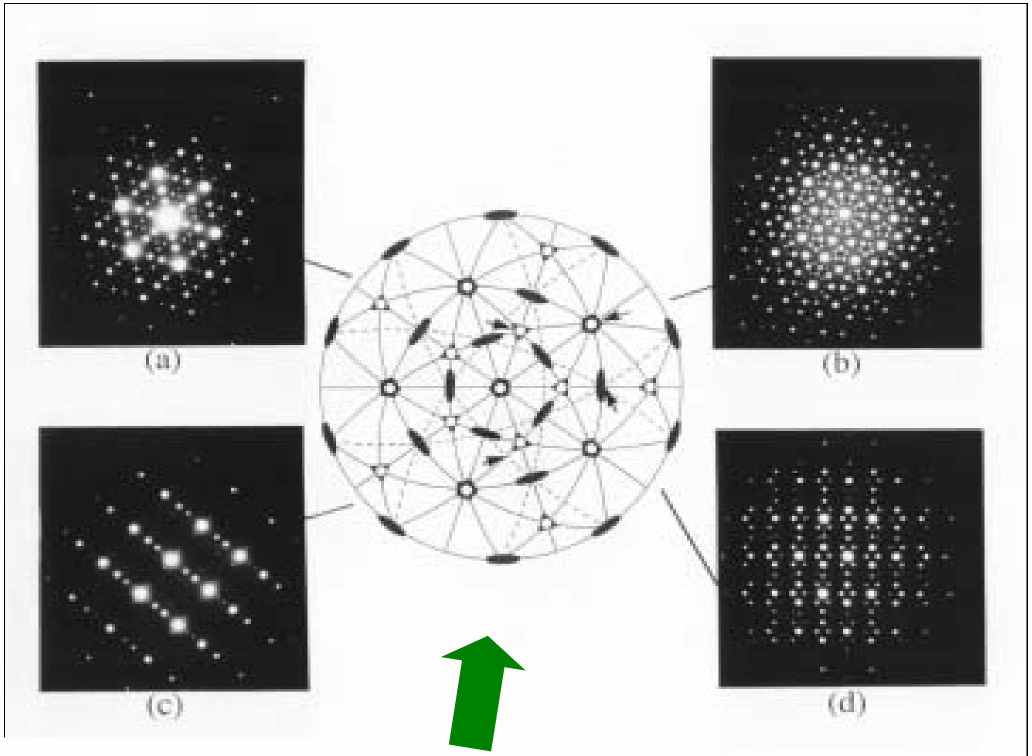
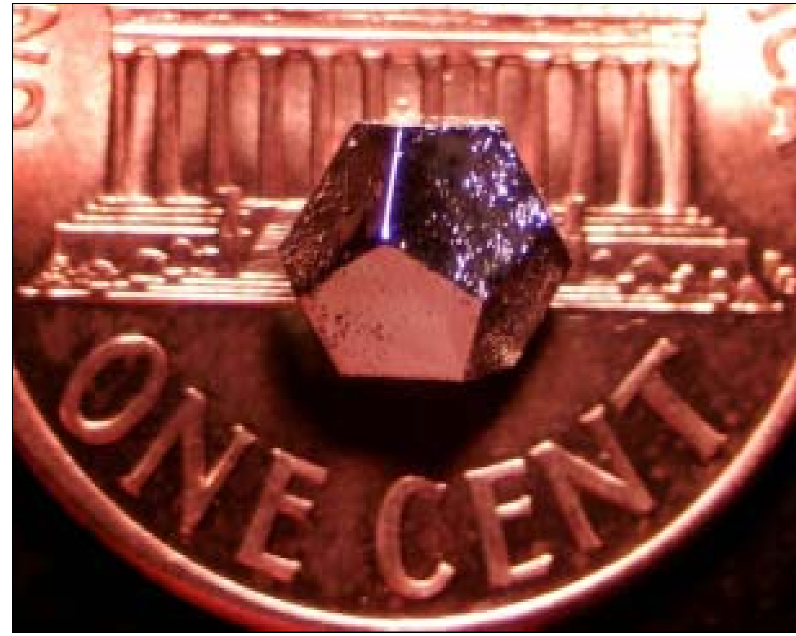
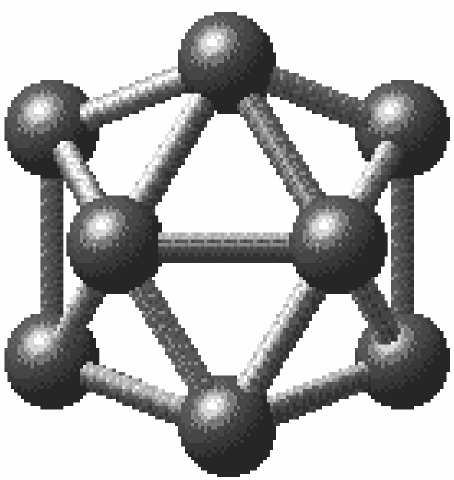
Icosahedral Quasicrystals (i-phase)



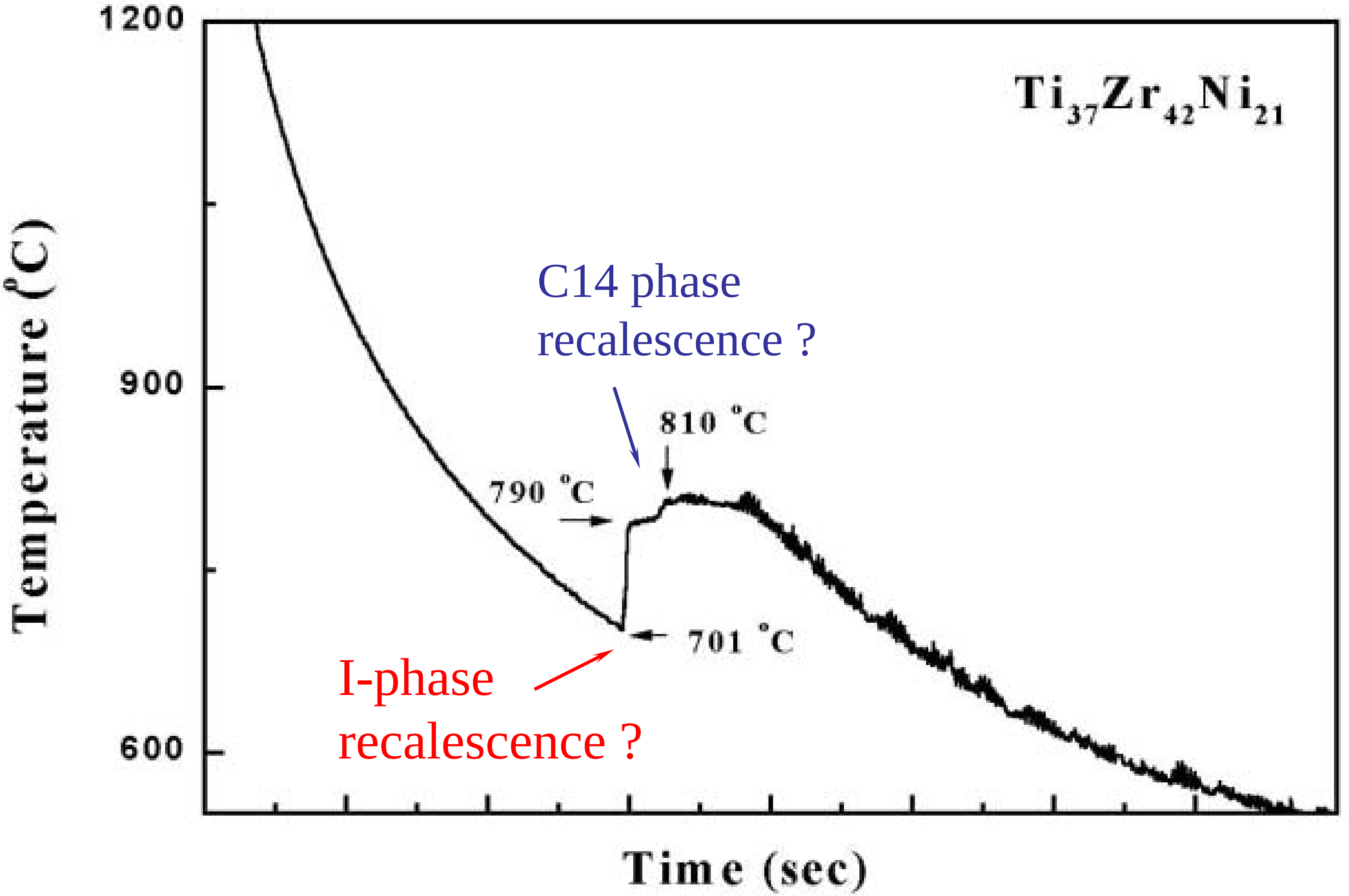


i(HoMgZn) - I. R. Fisher and P. C. Canfield

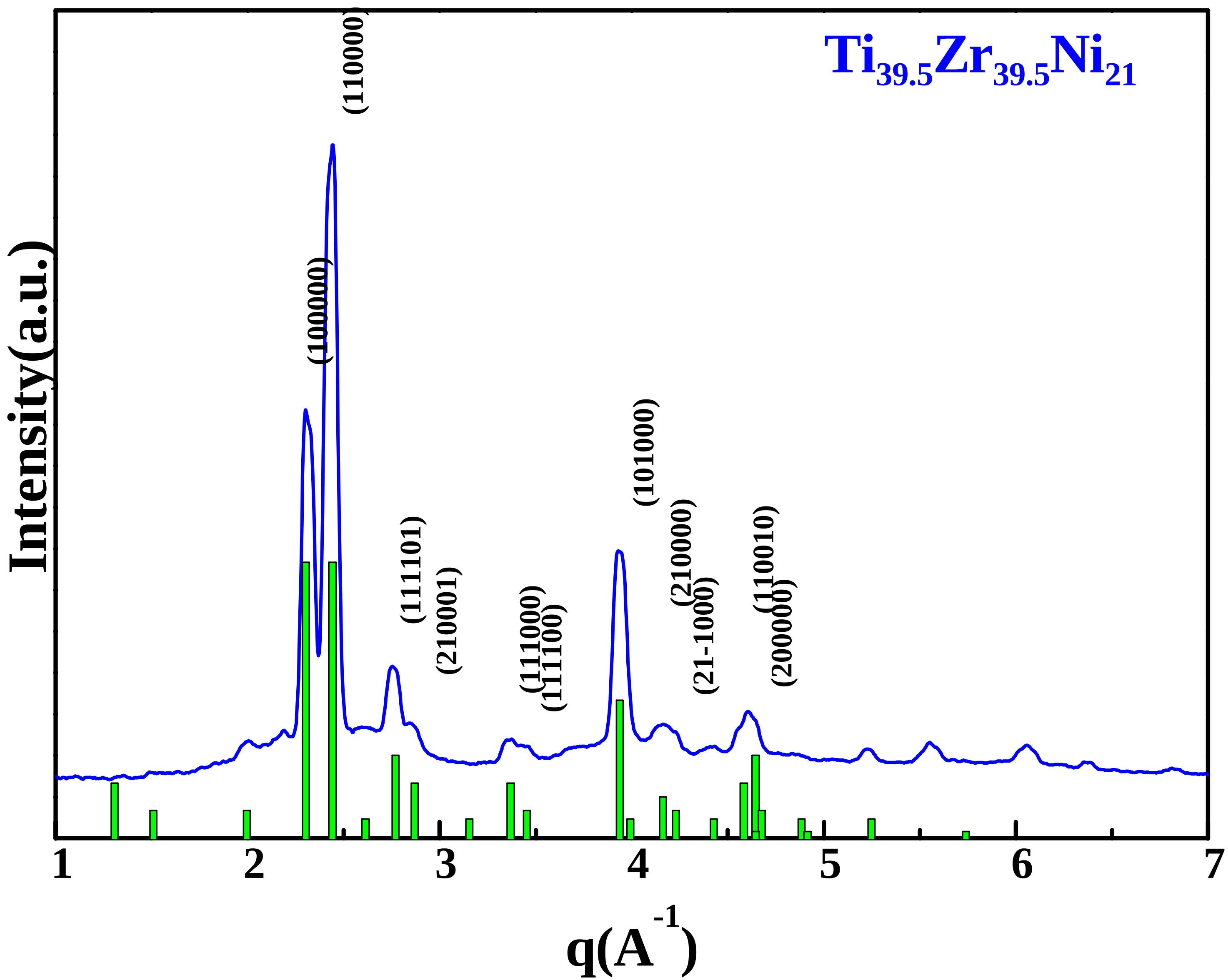


- Non-crystallographic symmetry
- Quasiperiodic translational order
- Local and extended icosahedral symmetry

ESL Undercooling Studies

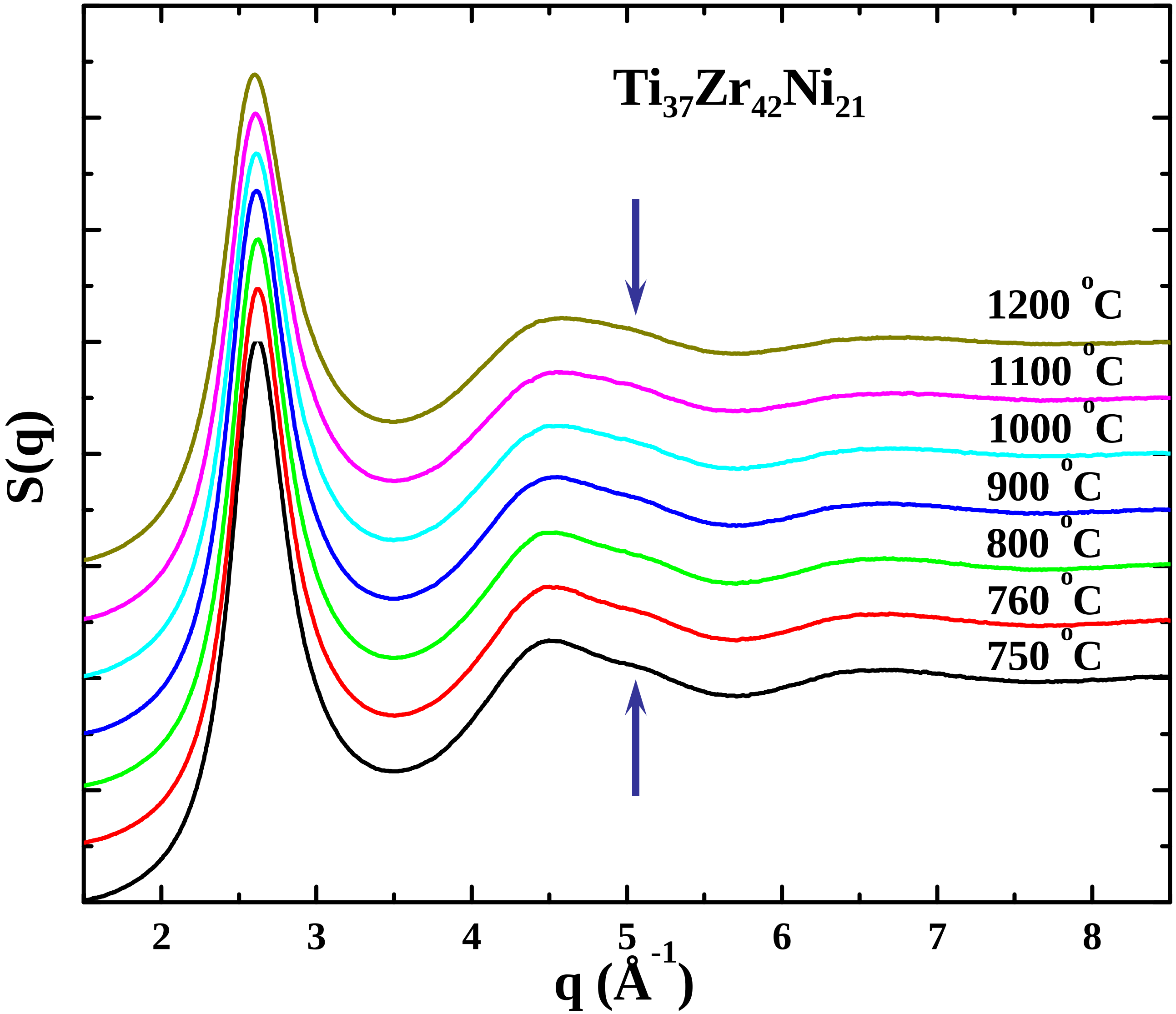


BESL of Liquid During Recalescence



Confirm that metastable i-phase is primary crystallizing phase

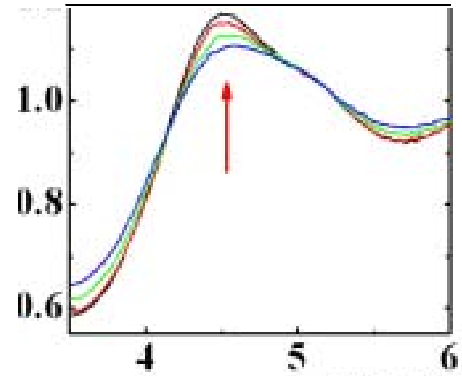
BESL Measurements of S(q) for $Ti_{37}Ni_{42}Ni_{21}$



Increased order with decreasing temperature

Coordination number is 12 ± 1

growing icosahedral order



*K. F.Kelton, A. K. Gangopadhyay, G. W. Lee, R. W. Hyers, R. J. Rathz, J. Rogers, M. B. Robinson, D. Robinson, Phys. Rev. Lett, **90**, 195504 (2003).*

Summary for TiZrNi

- Metastable i-phase forms in preference to equilibrium C14 phase
  - ⇒ i-phase has lower nucleation barrier
  - ⇒ Short-range order in liquid is icosahedral, not C14 tetrahedral type

- X-ray structural data indicate growing short-range order with increasing undercooling (shoulder)

- ***Taken together these indicate ISRO in TiZrNi liquid***

Rapid PDF Studies of Undercooled Liquid Si

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(b) Washington University St Louis, MO 63130 USA

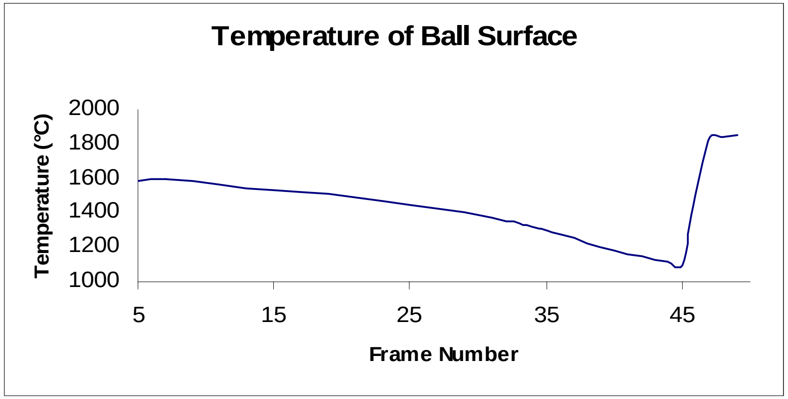
(c) NASA, George C Marshall Space Flight Ctr, Huntsville, AL 35812 USA

Why Study Levitated Liquid Si?

- Many discrepancies appear in the literature in relation to the properties of undercooled Si liquids, particularly concerning coordination numbers.

- Studies may provide useful insight into homogeneous crystallization of materials.

Heating/Cooling Cycle

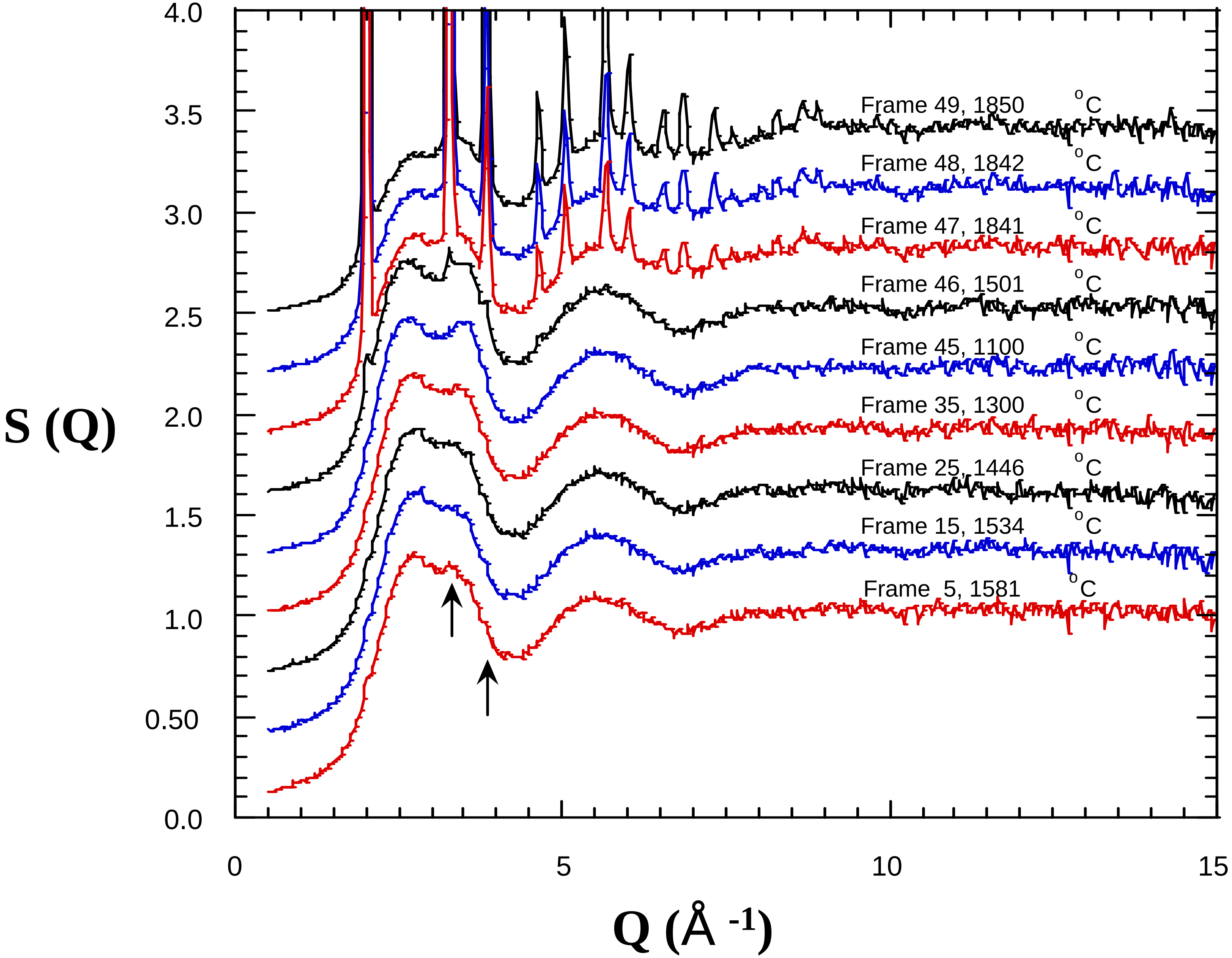


Frame Number	Temperature (°C)
5	1581
7	1596
9	1580
11	1565
13	1547
15	1534
17	1522
19	1506
21	1487
23	1470
25	1446
27	1425
29	1403
31	1372
33	1337

Frame Number	Temperature (°C)
34	1320
35	1300
37	1251
39	1203
40	1182
41	1164
42	1146
43	1130
44	1115
45	1100
46	1501
47	1841
48	1842
49	1850

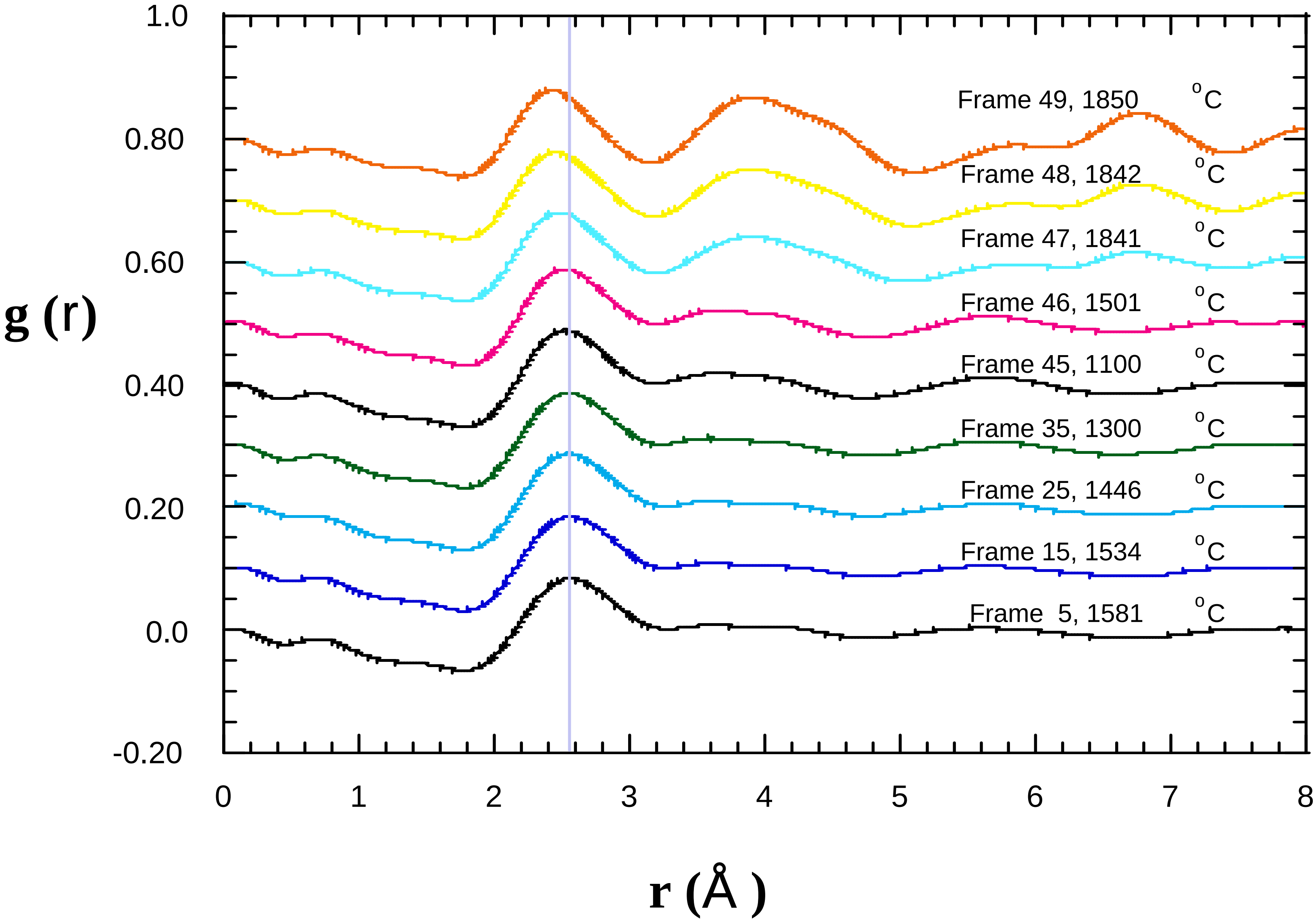
- Sample was heated above melting point (m.p. = 1414 °C) and held to ensure complete melting
- Sample was then cooled at controlled rate
- X-ray powder patterns were collected at one second intervals
- Recalescence occurred at 1100°C, yielding roughly 300 °C undercooling

S(q)



S (Q)
Q (Å -1)
Frame 49, 1850 °C
Frame 48, 1842 °C
Frame 47, 1841 °C
Frame 46, 1501 °C
Frame 45, 1100 °C
Frame 35, 1300 °C
Frame 25, 1446 °C
Frame 15, 1534 °C
Frame 5, 1581 °C
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.50
0.0
0
5
10
15

g(r)



g (r)
1.0
0.80
0.60
0.40
0.20
0.0
-0.20
0
1
2
3
4
5
6
7
8
r (Å)
Frame 49, 1850 °C
Frame 48, 1842 °C
Frame 47, 1841 °C
Frame 46, 1501 °C
Frame 45, 1100 °C
Frame 35, 1300 °C
Frame 25, 1446 °C
Frame 15, 1534 °C
Frame 5, 1581 °C

GENERAL SCIENTIFIC AND TECHNICAL GOALS

The ability to measure structures and kinetics at high temperatures while avoiding problems of sample holder contamination will enable fundamental studies of:

- short and medium range order in liquids above and below the liquidus temperature
- phase-transformation kinetics and their effect on materials processing
- metastable phase detection and structures
- aqueous and biological systems

Areas of technological importance that will benefit from the BESL facility include:

- fast and reliable phase-diagram determination, particularly of high-temperature materials,
- identification of solidification pathways, including the formation of transient metastable phases,
- *in-situ* studies of phase evolution during reactive phase transformation, such as the incorporation of oxygen in high-Tc superconductors,
- *in-situ* study of catalytic processes, of importance in sensor applications for example.

Current Status and Next Steps

- Building a User Community
  - Workshop in February generated significant interest
- Workshop report submitted to NASA calling for a chamber to be permanently based in the MUCAT Sector
- Always looking for good science!