High Temperature Resonant Ultrasound Spectroscopy (RUS) Methods

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Outline

- Introduction
- Potentials of RUS methods in high-temperature physics
- Design and fabrication of high temperature direct contact RUS probe
- Current and future research
- Elastic constant measurements of thermoelectric (TE) materials
- Summary

Introduction I:
Modulus of Linear Elasticity

Strain: $\epsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i})$

Generalized Hooke's Law: $\sigma_{ij} = c_{ijkl} \epsilon_{kl}$

Reduced index form: $11 \rightarrow 1, 22 \rightarrow 2, 33 \rightarrow 3, 23 \rightarrow 4, 31 \rightarrow 5, 12 \rightarrow 6$

Isotropic materials: $c_{11,} c_{44}$

Introduction II:
Methods for Measuring Elastic Constants

Mechanical methods for measuring elastic constants:

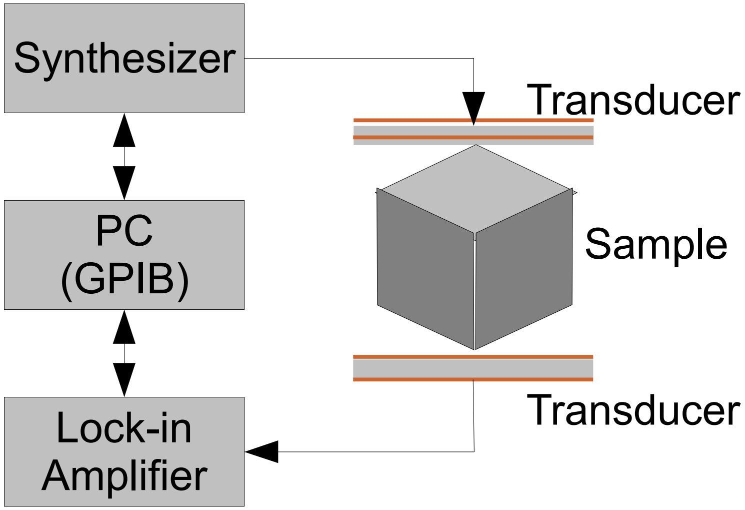
- Quasi-static method
- Pulse-echo method $v = \sqrt{\frac{C_{ij}}{\rho}}$
- Resonance method
  - Natural frequencies depend on geometry, density, elastic constants

Introduction to Resonant Ultrasound Spectroscopy (RUS)

RUS technique uses resonance spectra of elastic bodies to infer material properties such as elastic moduli

Typical RUS procedures:

- Prepare the specimen – cutting & polishing
- Get the dimensions, shape, density, crystallographic orientation etc.
- Drive the specimen over a swept frequency range
- Identify the resonance peaks
- Adjust the material parameters (elastic constants) to best fit the experimental resonance spectrum in the least square sense



Synthesizer

PC (GPIB)

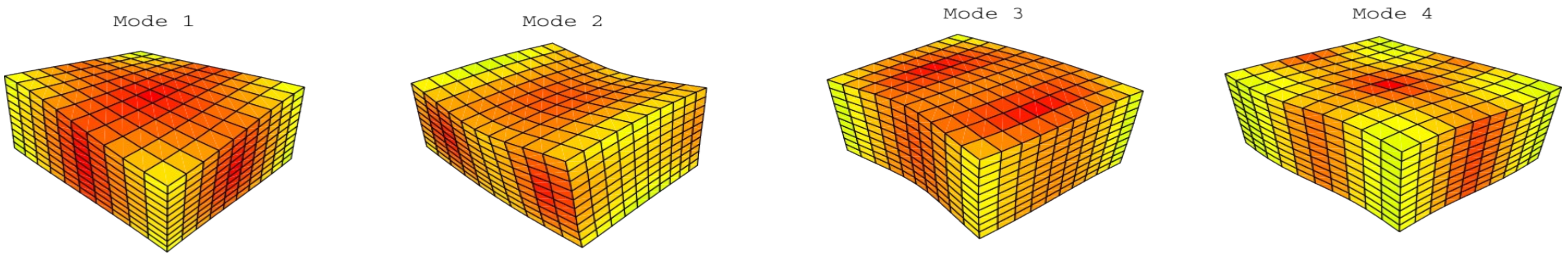
Lock-in Amplifier

Transducer

Sample

Transducer

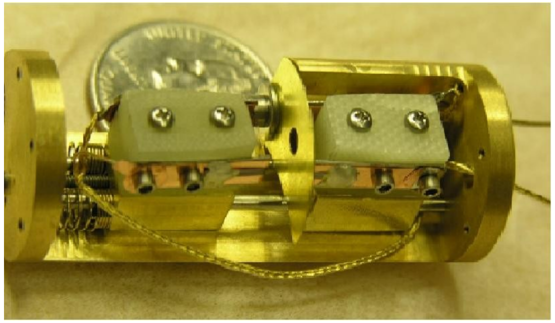
RUS system diagram



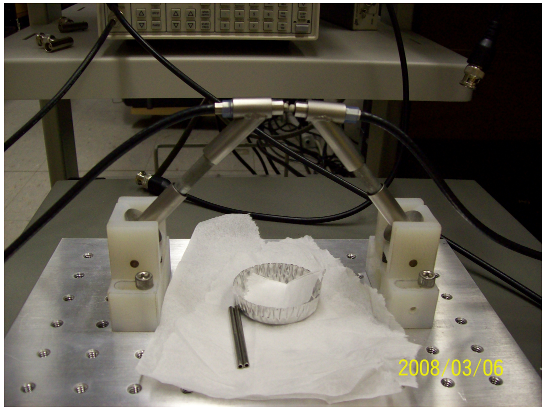
Four normal modes of a parallelepiped

Advantages Over Other Ultrasonic Methods

- Obtain a full elastic tensor in one frequency sweep
- Highest accuracy
- Fast (partly due to modern computing power)
- No bond between the sample and the transducer
- Works well with small (sub-mm) & low-symmetry crystals

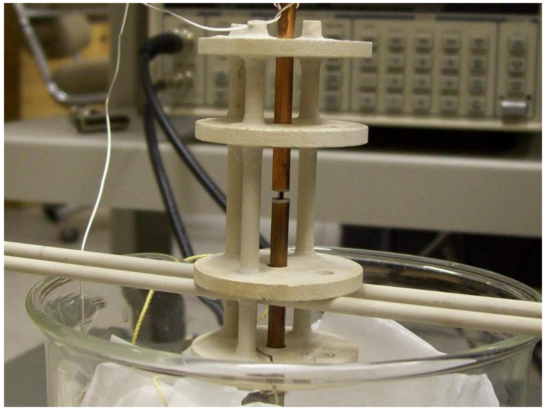


RUS cell with
PVDF transducers



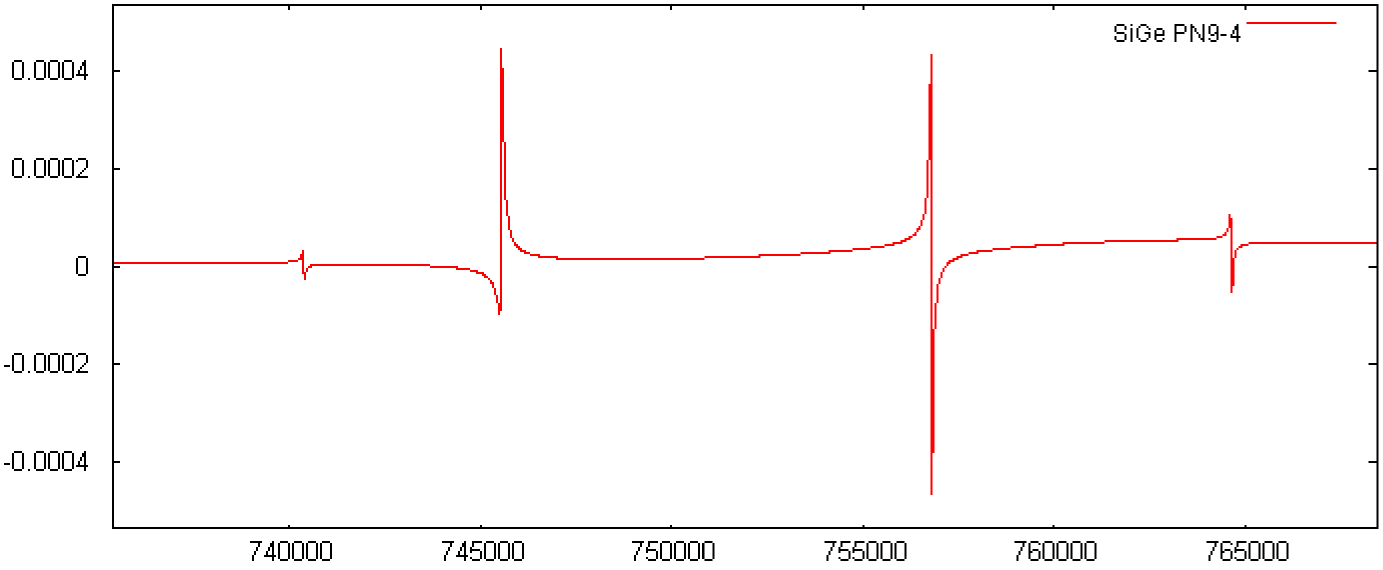
DRS Q9000 RUS
System transducers

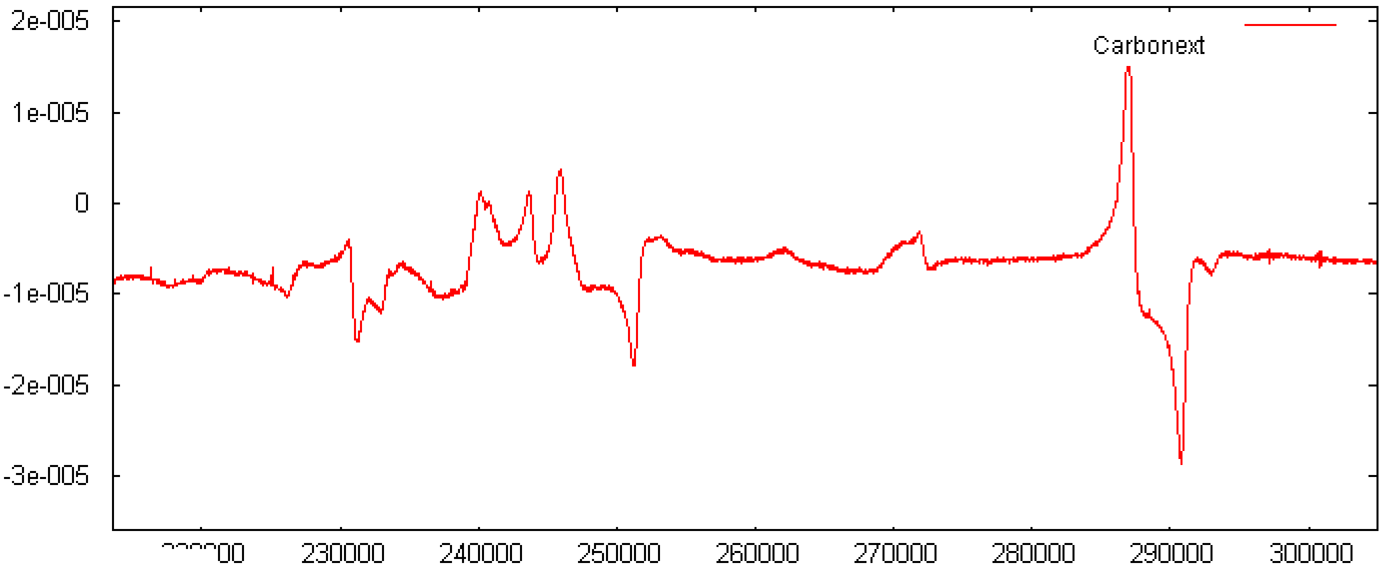
Transducer on a stage



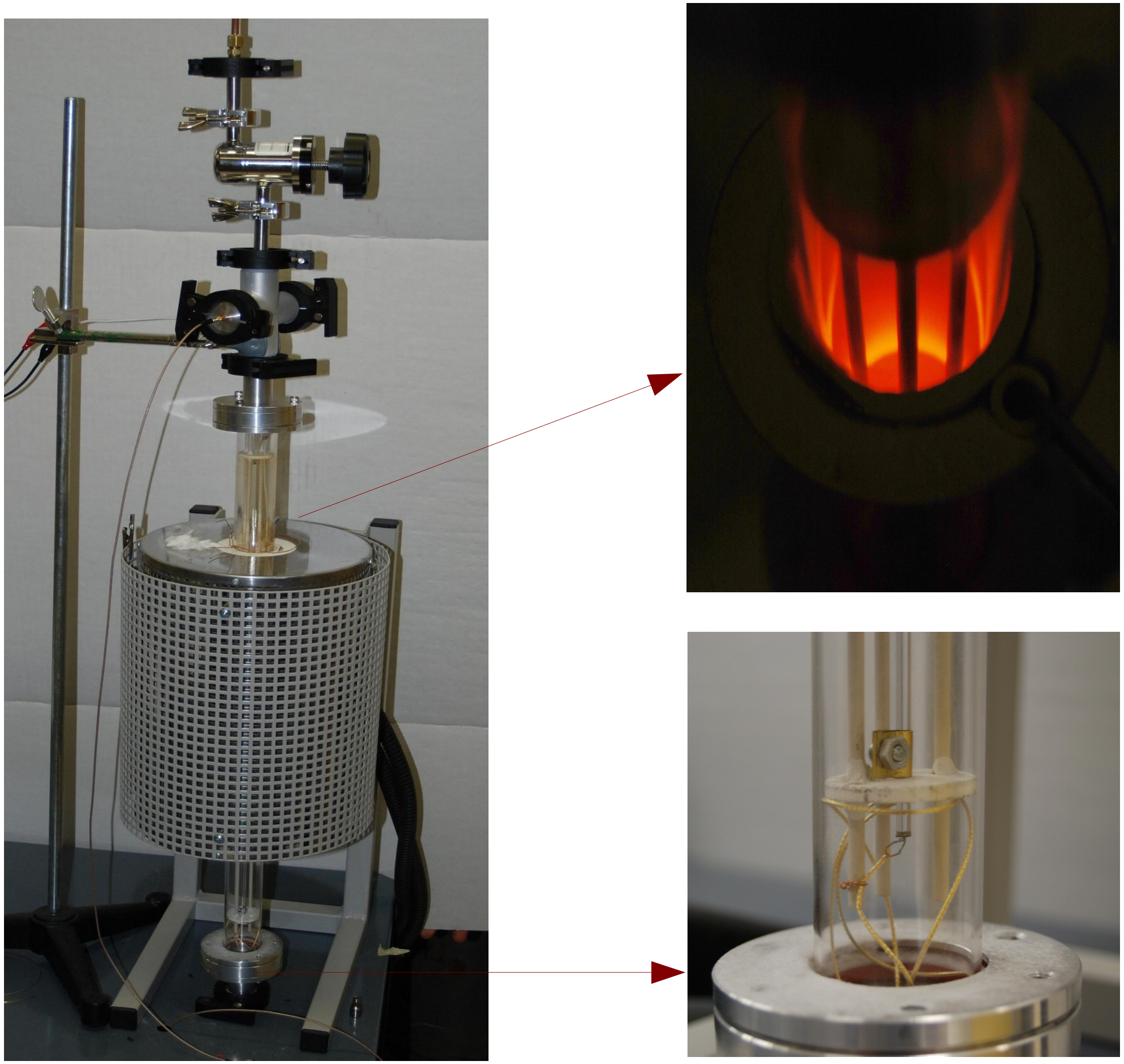
Mid-temperature
direct-contact transducers

Representative Resonance Spectra
Ideal vs. bad





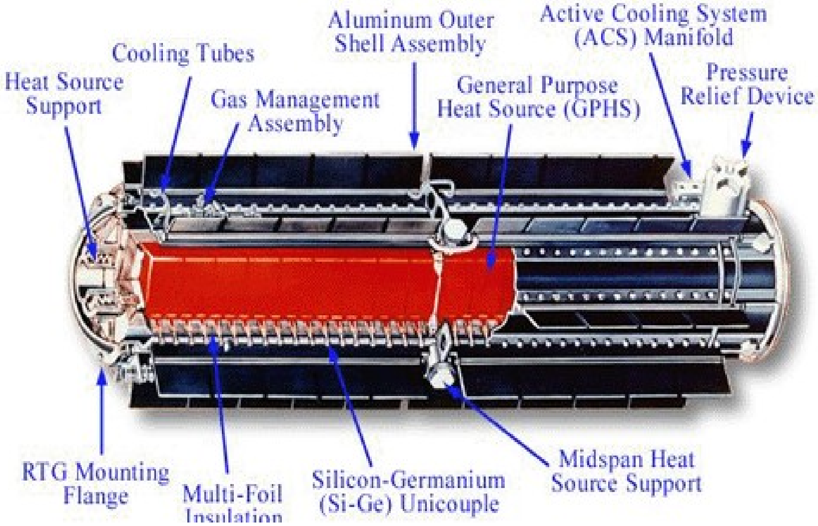
High Temperature RUS System



High-temperature RUS system

Potentials of RUS Methods in High-temperature Physics

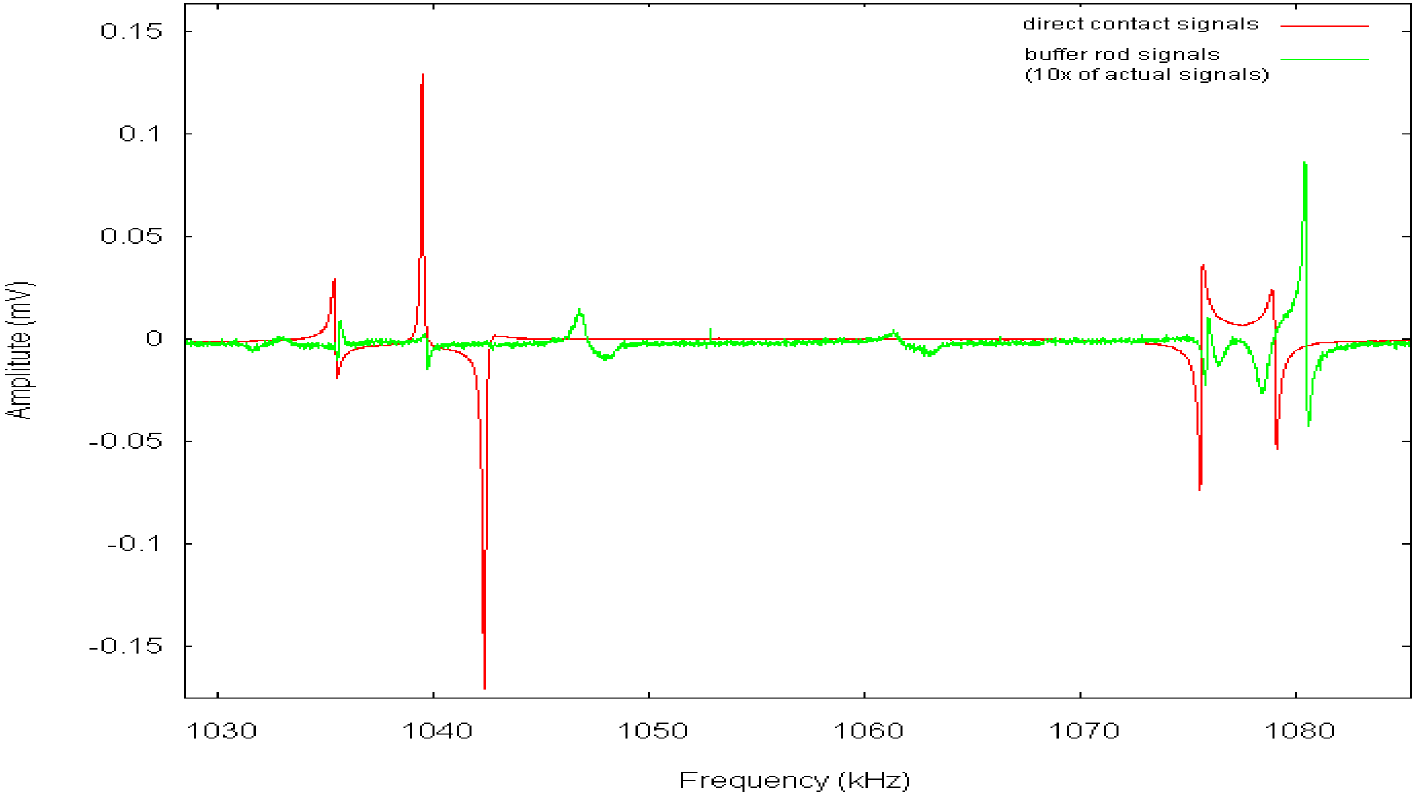
- RUS has been valuable for the study of condensed matter physics and materials science
- Temperature-dependent elastic properties of materials are sensitive probe into their atomic environment
- By extending the RUS method to high temperatures, a variety of fields can be investigated:
  - Thermoelectric materials – SiGe, Zintl phases
  - Structural phase transitions in crystals
  - Glass transitions in bulk metallic glasses (BMGs)
  - Elastic properties & dissipation of novel materials
  - Non-destructive evaluations or characterization of materials (new ceramics)



Cutaway view of Cassini's RTGs
* Courtesy of Jet Propulsion Laboratory

Challenges of RUS Measurements at Very High Temperatures

- Most commercial transducers are not designed to operate above 200C
- Electrical connection is non-trivial
- Oxidation – low flow 95/5 argon/hydrogen
- Temperature control challenges – convection of low flow inert gas
- Unwanted rod resonance from buffer rods

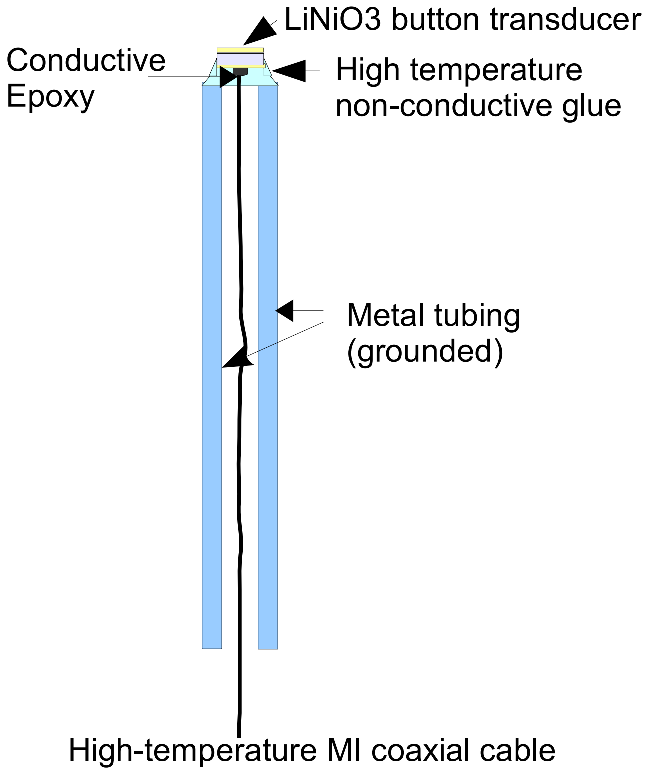


Piezoelectric Materials Selection and Direct Contact RUS Probe Diagram

Material	**Curie point(°C)**
PVDF	165
Ceramics (PZT)	200~500
Crystal quartz	573
Tourmaline	>= 900
GaPO4	970
Lithium Niobate	1150
Aluminium Nitride	1200
PiezoStar®(Kistler)	>1300

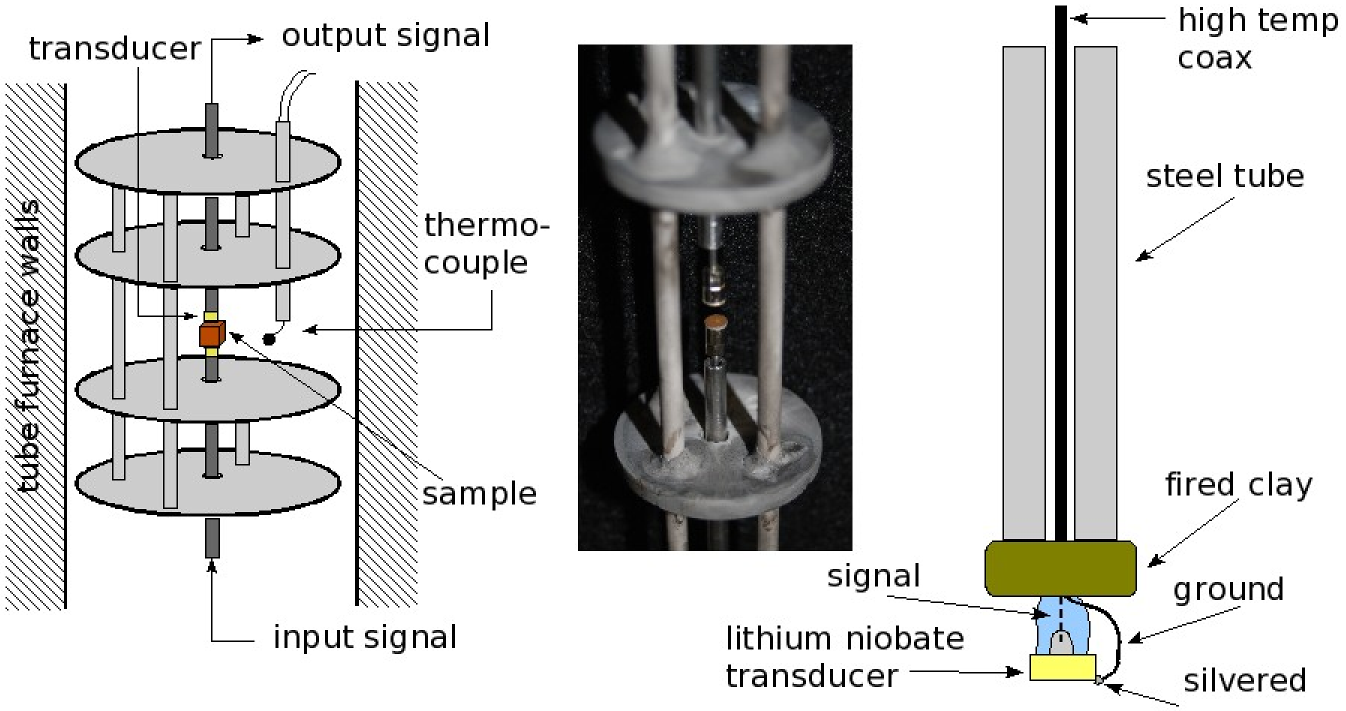
Lithium Niobate is good candidate!

Piezoelectric materials &
their Curie points



Direct-contact high-temp
RUS probe

Direct Contact RUS Probe

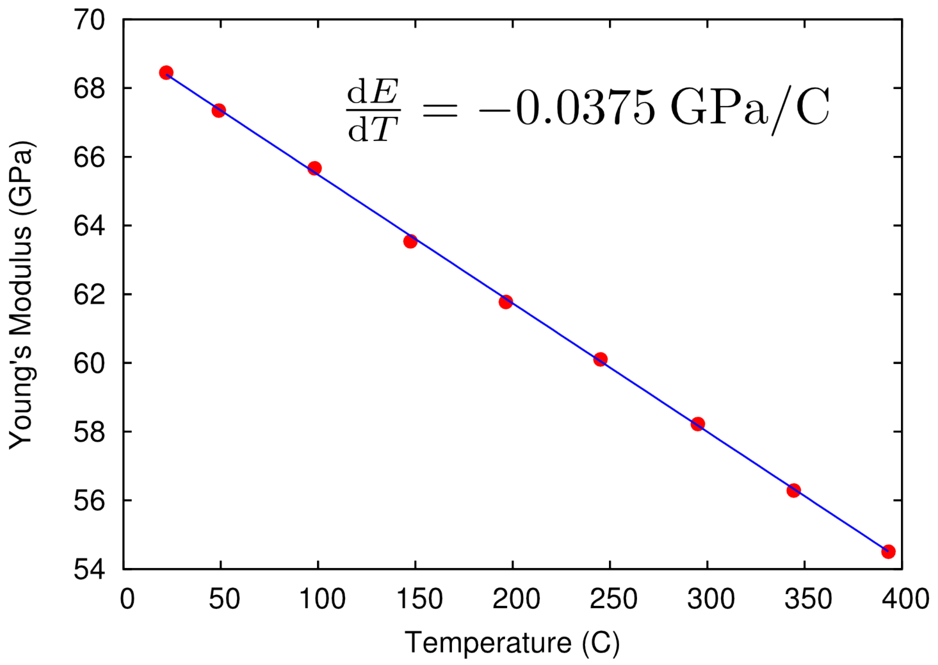


(a) (b)

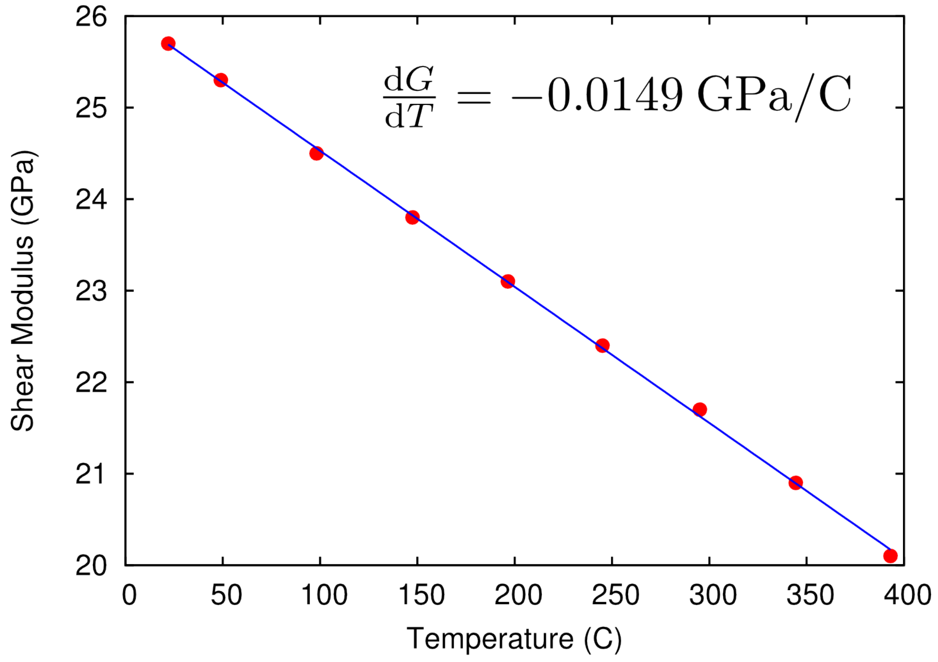
Past & Current Research With the High-temperature RUS System

- Test run the system with materials of well known properties – quartz, aluminum, etc.
- Elastic properties of a novel piezoelectric material, kepertite at high temperatures
- Charge-order phase transitions in transition metal oxides
- Elastic properties of thermoelectric materials
  - Zintl phases
  - SiGe (doped)
- Glass phase transition of bulk metallic glasses (BMGs)

High temperature RUS System Test Run: Temperature Dependent Elastic Constants of Aluminum



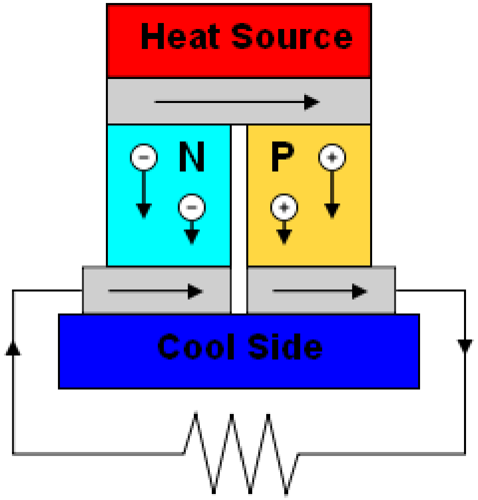
Temperature dependence of Young's modulus



Temperature dependence of shear modulus

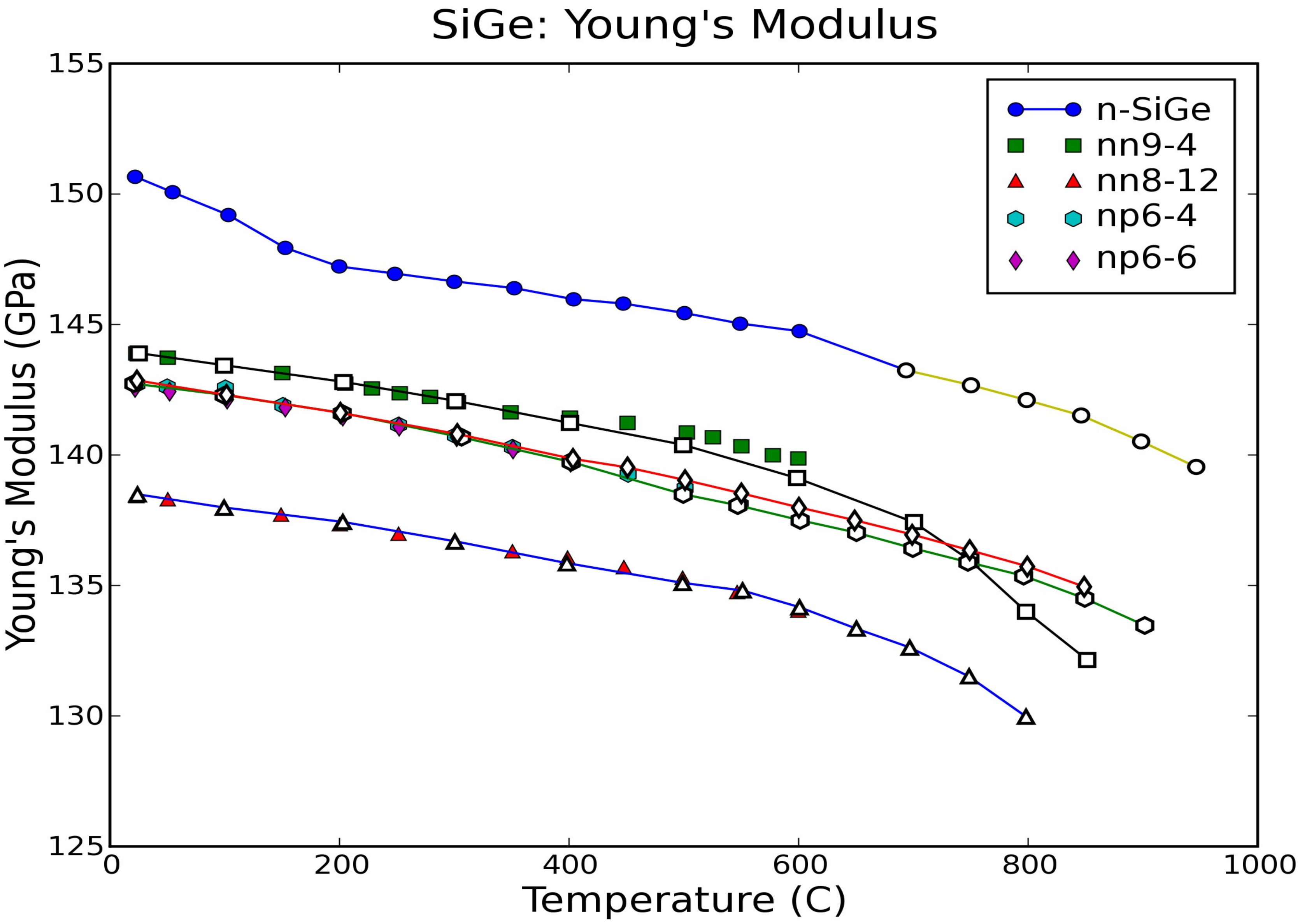
Thermoelectric (TE) Materials

- A little background - Seebeck effect (1821)
- Waste heat recovery for power generation – contributes to energy independence
- High performance TE materials for power generation on deep-space probes
- SiGe alloy is an excellent TE material for high temperature applications
- Mechanical properties required before their actual applications in high temperature harsh environment

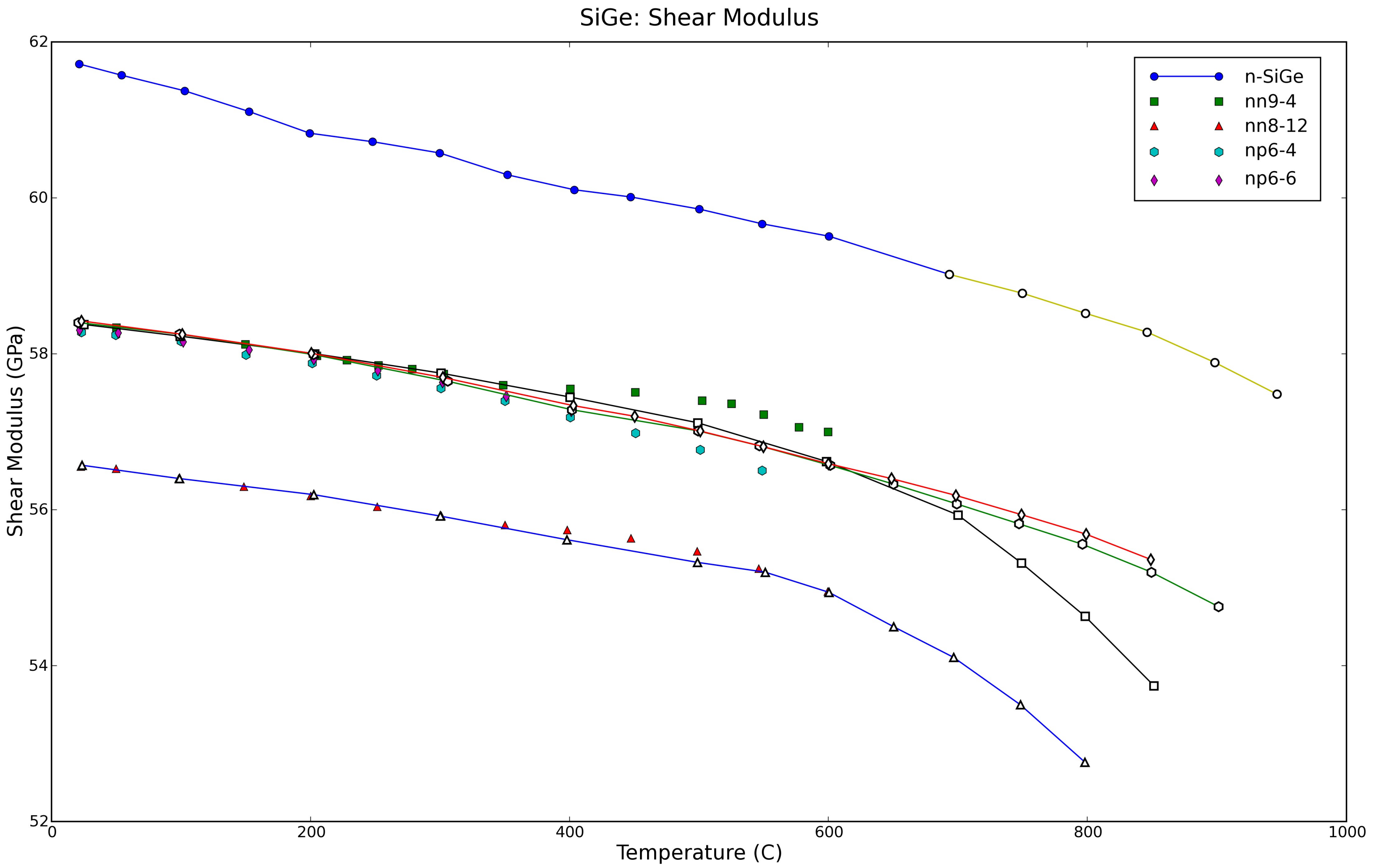


From wikipedia

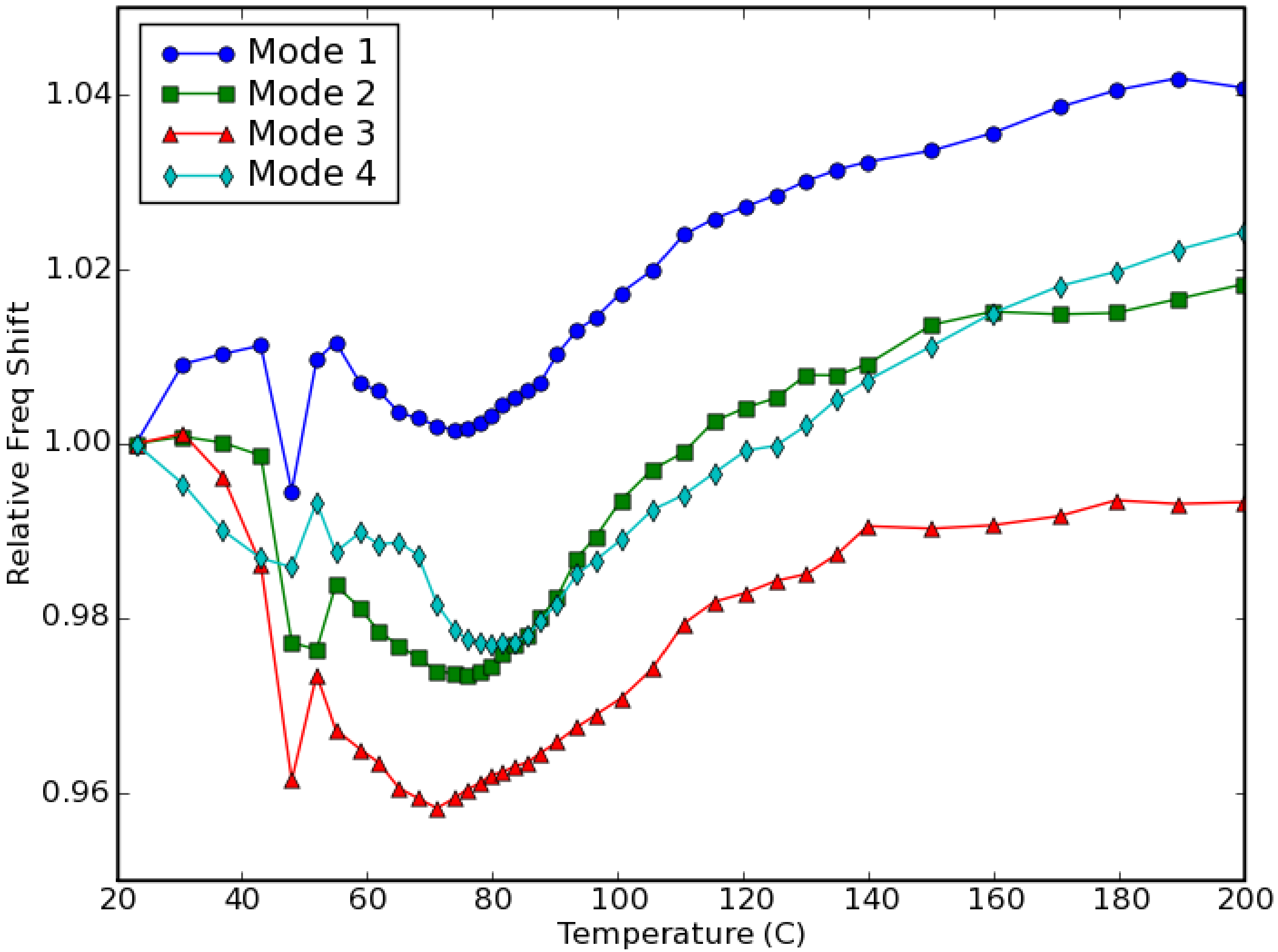
Young's Moduli for Four SiGe Materials: NN8-4, NN9-4, PN6-4,PN6-6



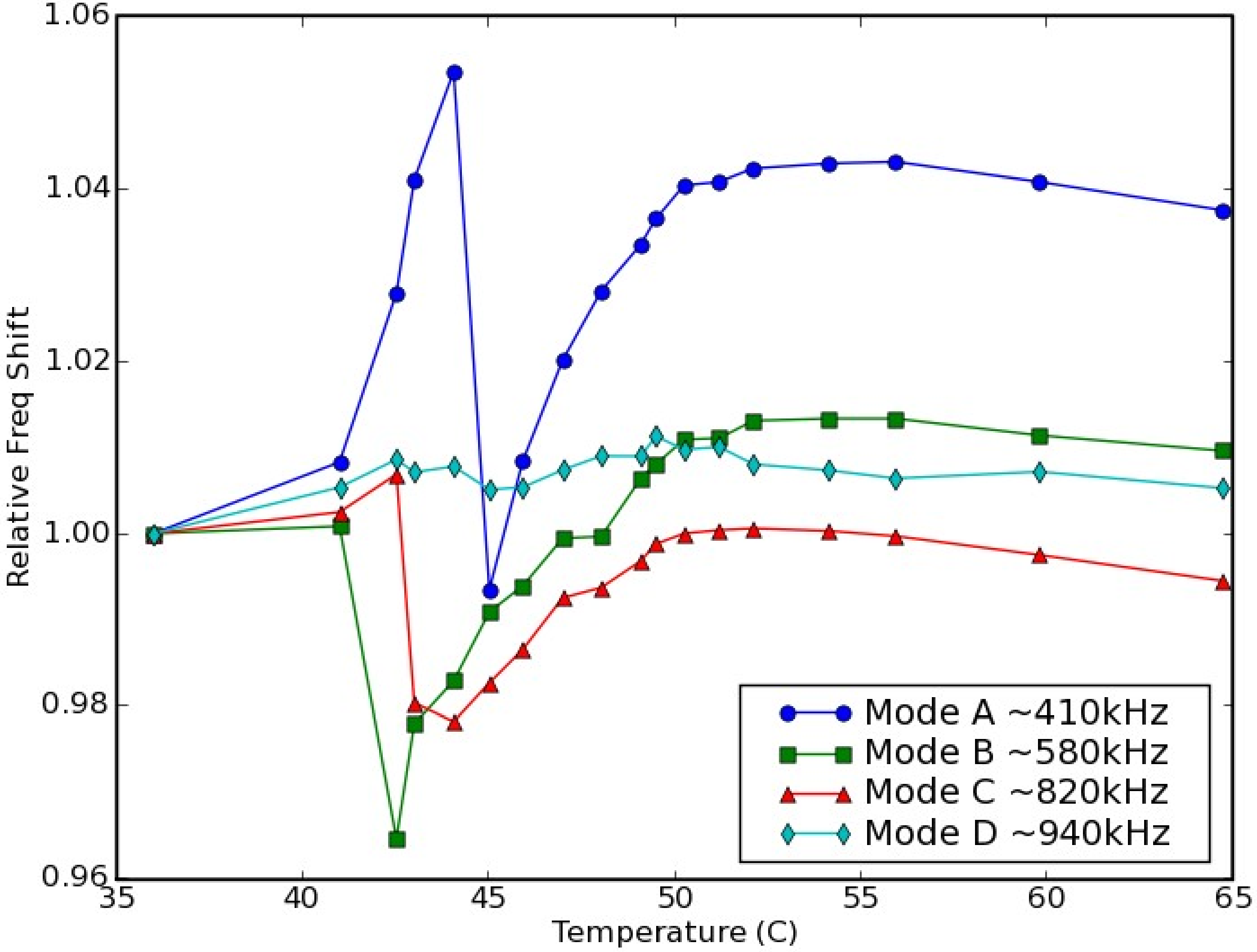
Shear Moduli for Four SiGe Materials: NN8-4, NN9-4, PN6-4,PN6-6



Phase Transition Studies in $LuFe_2O_4$ I:



Phase Transition Studies in $LuFe_2O_4$ II:



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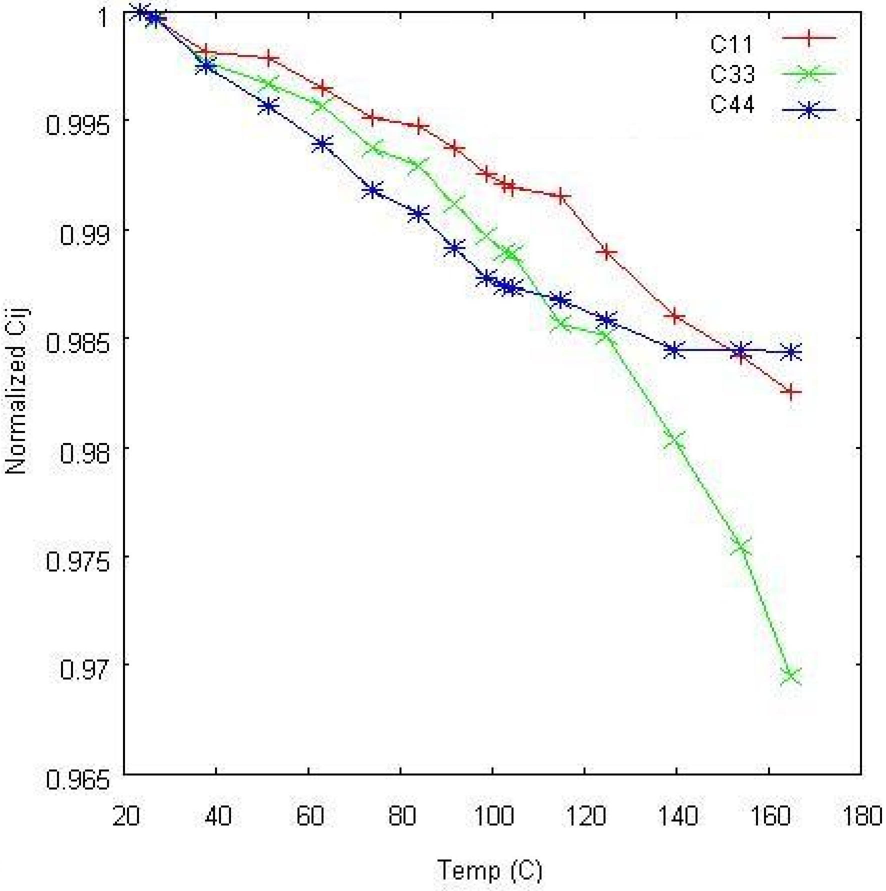
The End

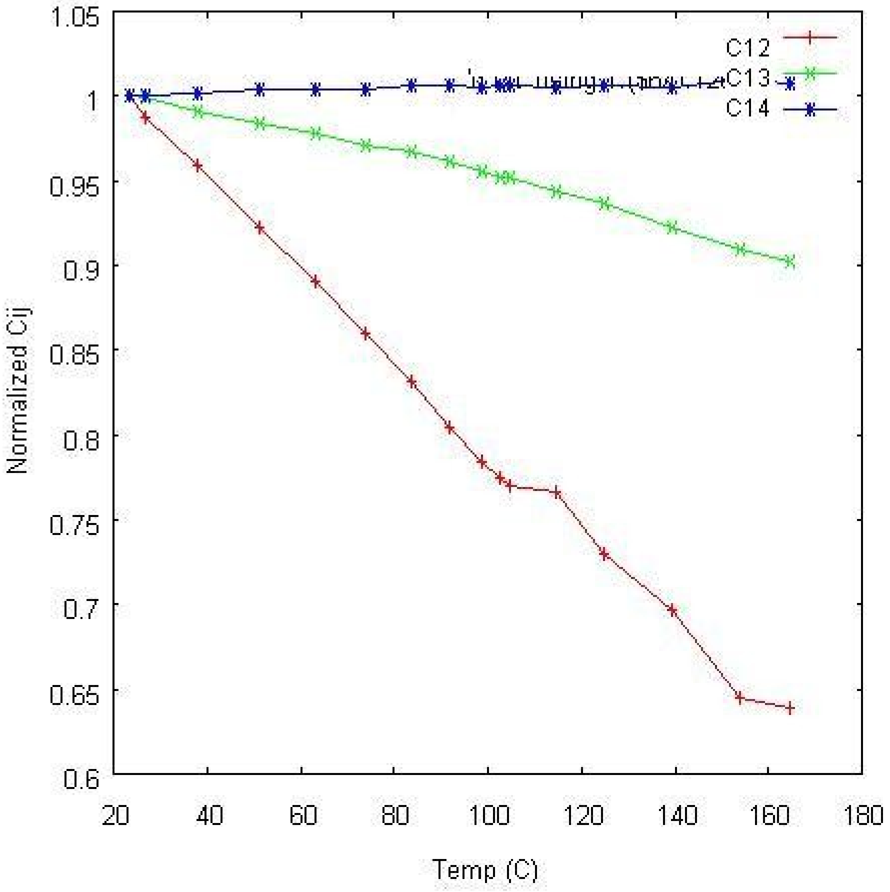
Thank you very much!

any questions?

High-temperature RUS Application Example II: Elastic Constants vs. Temp. for Quartz

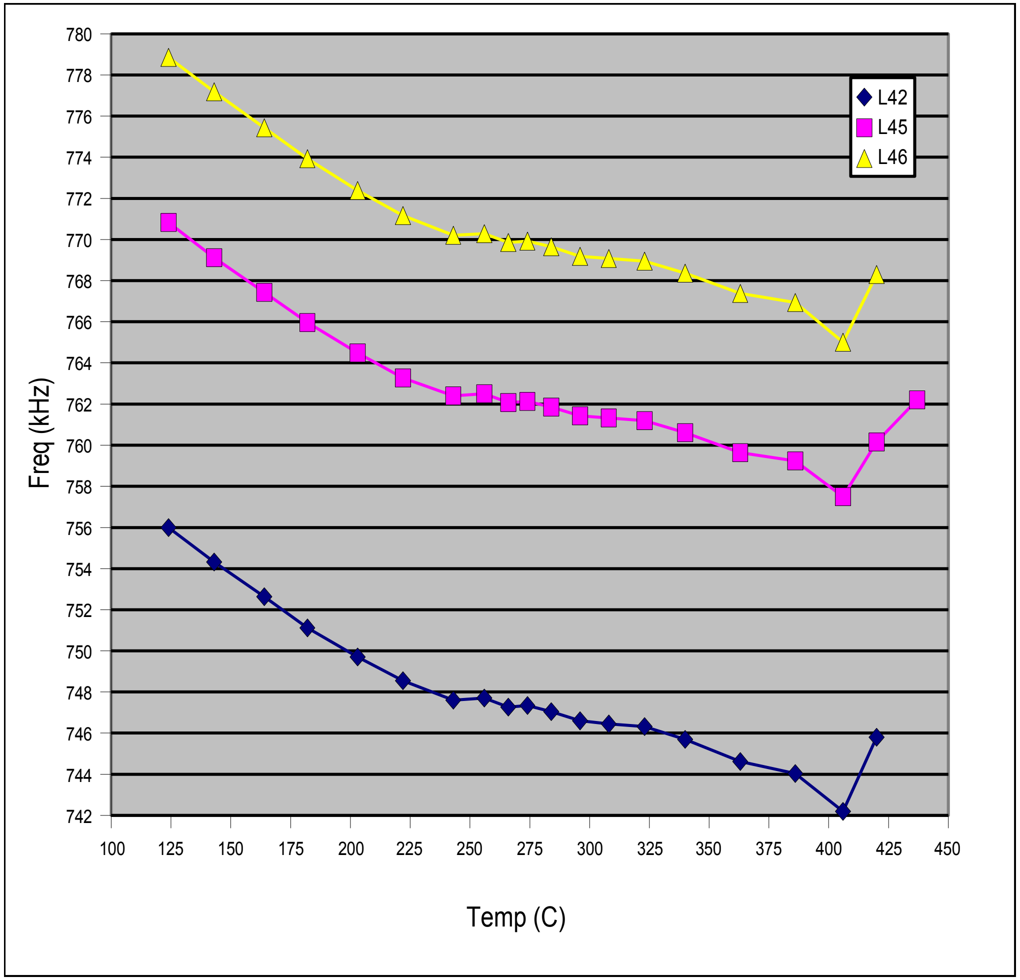


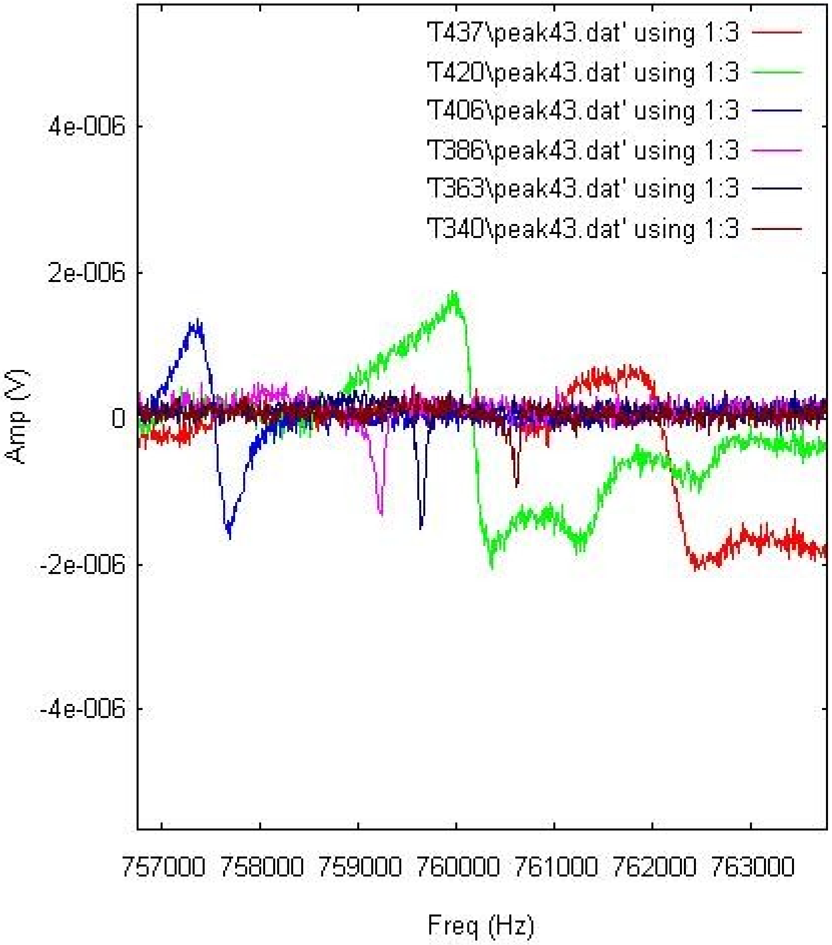
C11, C33, C44
vs. temperature



C12, C13, C14
vs. temperature

High-temperature RUS Application Example III: Phase Transition for BMG ($Zr_{50}Cu_{40}Al_{10}$)





Mode 42, 45 & 46
vs. temperature

An example of mode shift vs.
temperature for mode 45