Experimental determination of the interatomic force constant in earth and planetary materials

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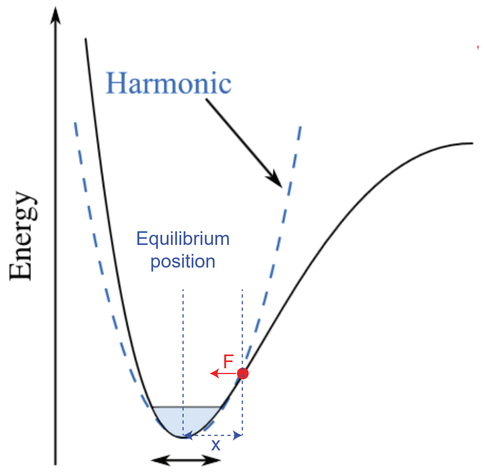
Abstract

Interatomic force constant is an essential physical parameter of condensed matter that determines numerous physical phenomena such as phase transition, thermodynamics, elasticity and isotopic behaviors. Although modern computational algorithms are capable of computing interatomic force constant under theoretical assumptions, an accurate experimental determination of interatomic force constant is imperative, especially under extreme pressure and temperature conditions. In this study, we present our recent experiments that aim to constrain the interatomic force constants in various earth and planetary materials at relevant high pressure-temperature conditions. Our study utilizes multiple X-ray based-techniques that are available at 3rd generation synchrotron radiation sources. We discuss the application of our force constant measurement on the isotope fractionation behaviors of earth and planetary materials.

Background

Interatomic force constant is the physical quantity that describes the restoring force that exerts on an atom when the atom is displaced from its equilibrium position. In crystalline minearals, the interatomic force constant is closely related to the atomic vibrations around the equilibrium position.

Figure 1: Diagram showing the relationship between the restoring force (F) and the atomic vibrations around the equilibrium position (x). In classic mechanics, the interatomic force constant is defined as:
N = F/x



There are two different kinds of interatomic force constants, namely the resilience and the stiffness.
Resilience (Nr) is traditionally defined as:

$$N_r = \frac{k_B}{d\langle u^2\rangle/dT}$$

where k_B is the Boltzmann constant, $\langle u^2\rangle$ is the atomic mean square diplacement, which is measureable from single crsytal X-ray diffraction.

Stiffness (Ns) is defined as:

$$N_s = \int M\left(\frac{E}{\hbar}\right)^2 D(E)dE$$

where M is the atomic mass the isotope, ħ is the reduced Planck constant and D(E) is the partial phonon density of states. The stiffness has geochemical significance in determining the equilibrium isotope fractionation.

Acknowledgements

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Experiments: single crsytal diffraction at 13-BM-C

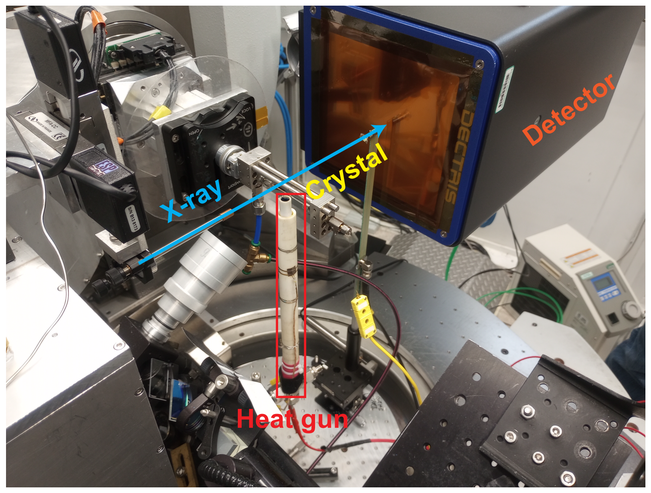


Figure 2: High temperature single crystal X-ray diffraction setup at APS 13-BM-C, tested with Pilatus 1M CdTe detector.

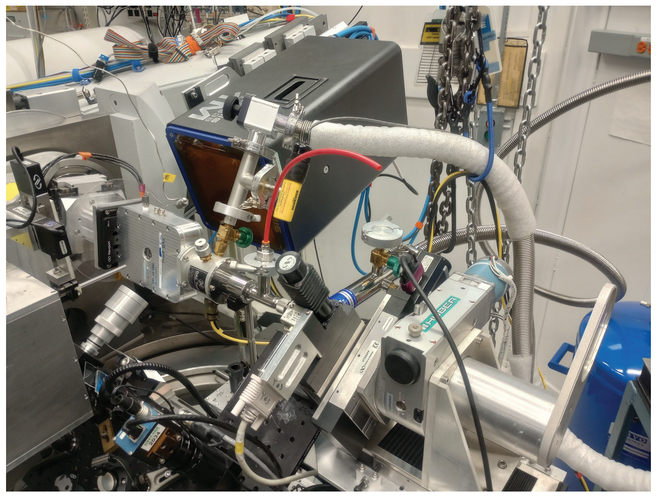


Figure 3: High pressure, low temperature single crystal diffraction setup at 13-BM-C, tested with the GSECARS cryostat.

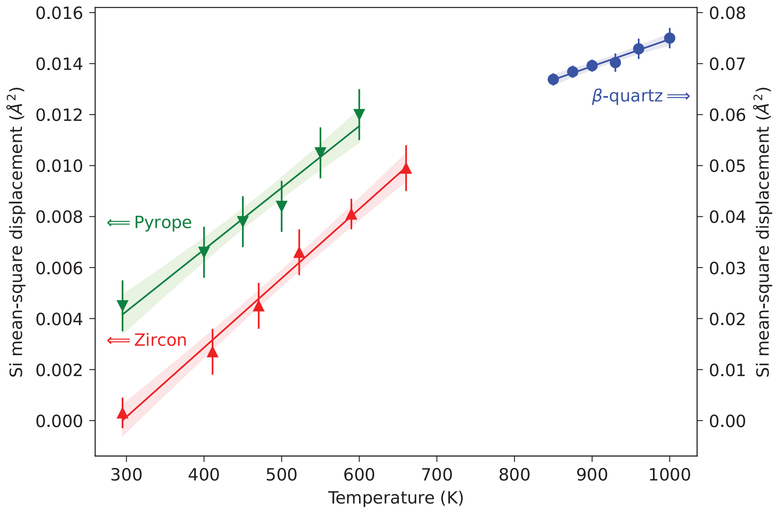


Figure 4: The Si atomic mean square displacement $\langle u^2\rangle$ as a function of temperature determined by the high-T XRD experiments from this study. Best linear fits and fitting error ranges are shown as the straight lines and shaded regions.

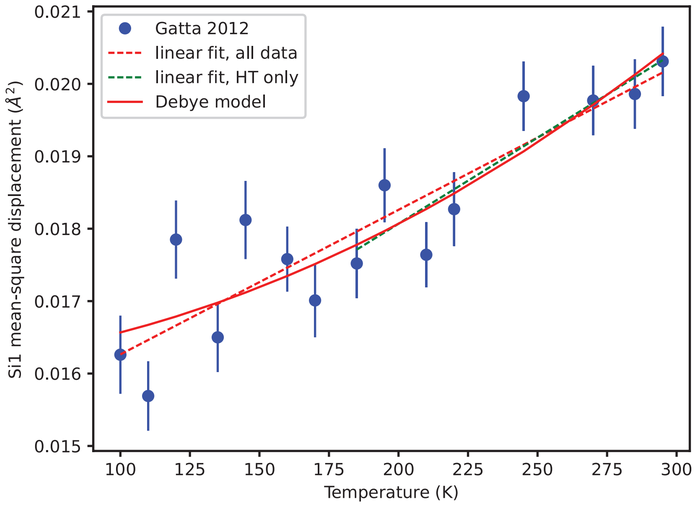
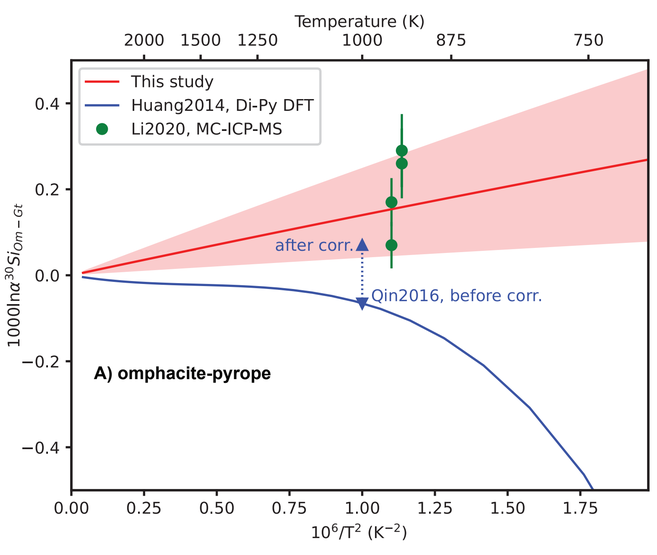


Figure 5: For low temperature data, our preliminary results suggest that a simple linear model would overestimate the Nr. Introducing a quantum phonon model (e.g., Debye) will be needed to fit the Si atomic mean square displacement $\langle u^2\rangle$ as a function of temperature in epidote.

Geochemical application



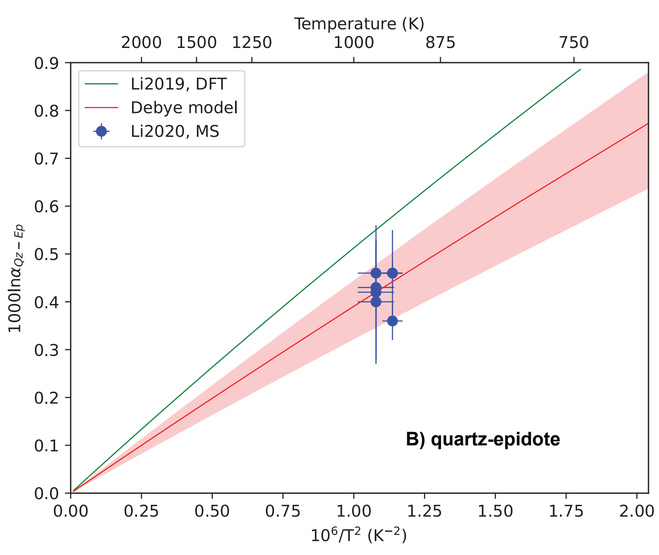


Figure 6: Equilibrium Si isotope fractionation between A) omphacite and pyrope and B) quartz and epidoite . Red curve: force constants approach. Red shaded region: uncertainty range of the force constants approach, determined from the distribution of the resiliences of the two minerals.