Pressure-induced Structural Modification in Framework Aluminosilicate

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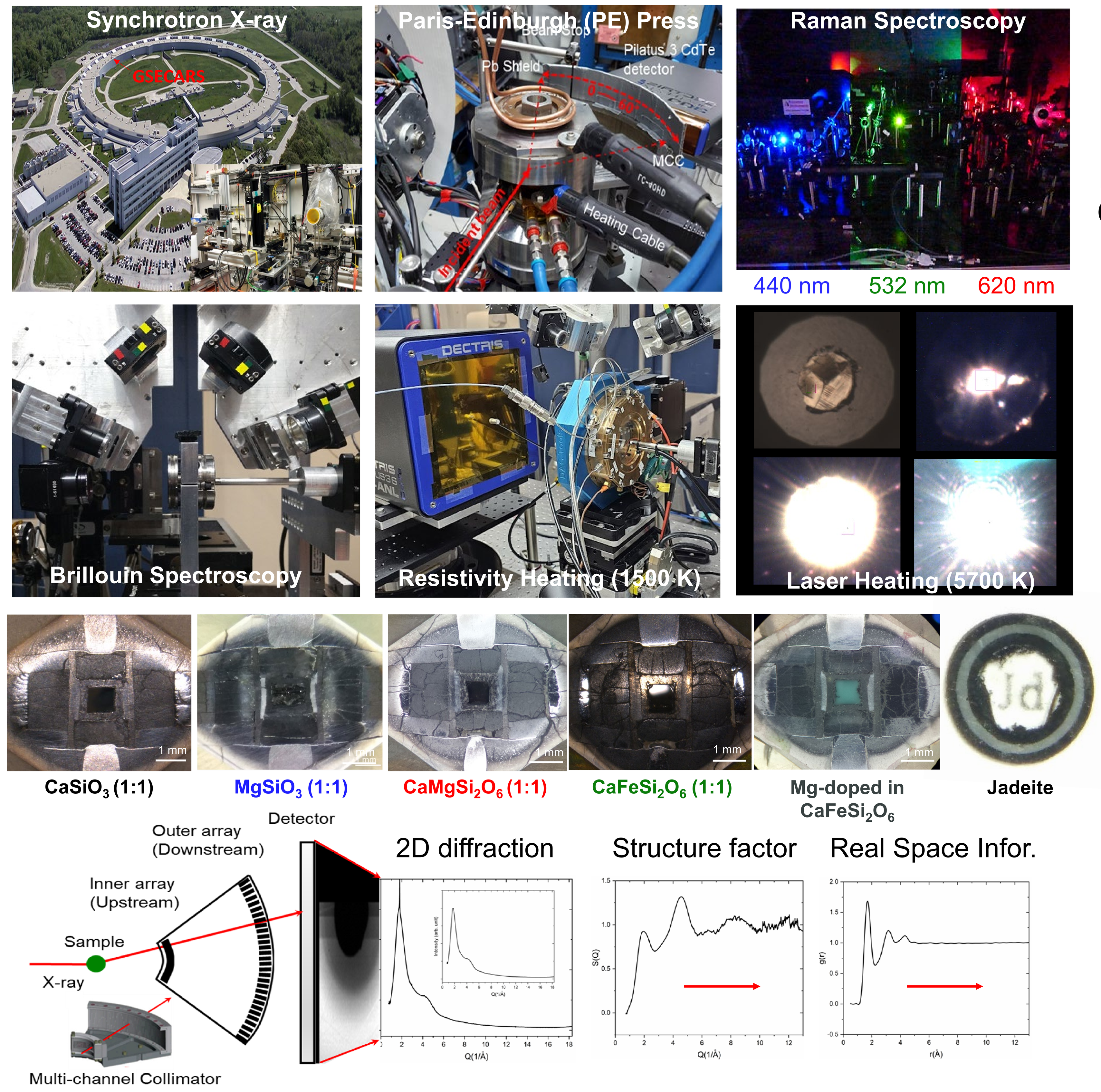
Background and Motivation

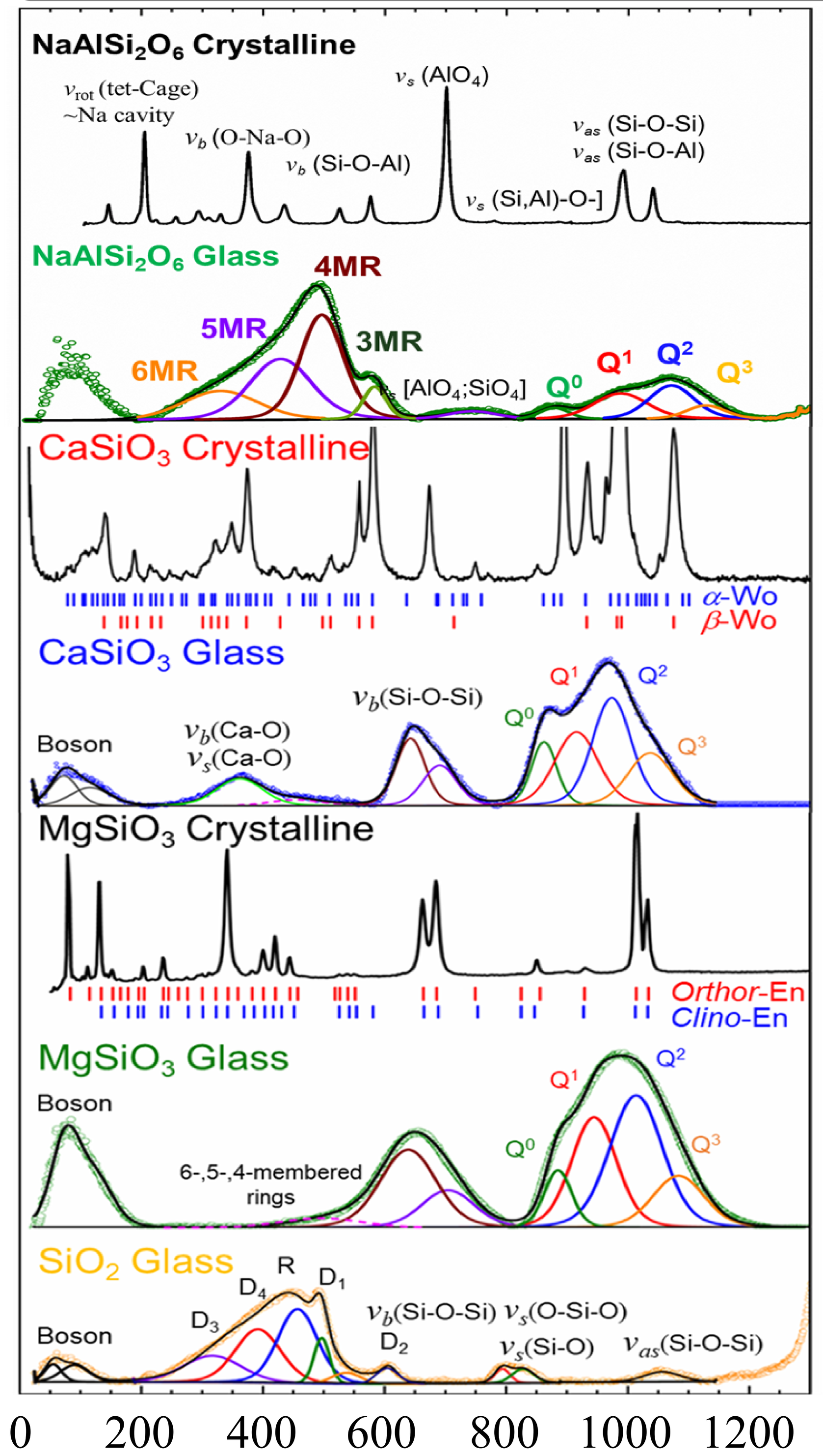
- Structure modification in silicate melts under pressure and temperature directly influence melt density in the Earth's interior, controlling the styling of melt migration. It is important to accurately understand how melt structure responds to pressure in order to predict properties like density, viscosity, and phase equilibria of magma within the Earth.
- Understanding the physical and chemical properties of silicate liquids and glasses requires studying their behaviors under high-pressure and high temperature. [1-4].
- However, little is known about the structures of silicates throughout the Earth's pressure regime because the high-temperature and pressure entail severe experimental difficulties.
- In order to understand the origin and significance of deep melts in Earth's interiors, effects of cation (Ca^{2+}, Mg^{2+}, $Al^{2+,3+}$) on silicate liquids and glass were investigated through studies of wollastonite (Wo), enstatite (En), Albite (Ab), and Jadeite (Jd) at extreme conditions. [5-6]
- Supercooled liquids (glasses) are of interest in elucidating structure of liquids under extreme pressures.
- An insight into the physical properties of the abovementioned samples gained through this study will expand our understanding of the behavior of silicate glasses under extreme conditions.

Instrumentation and Experimental Method



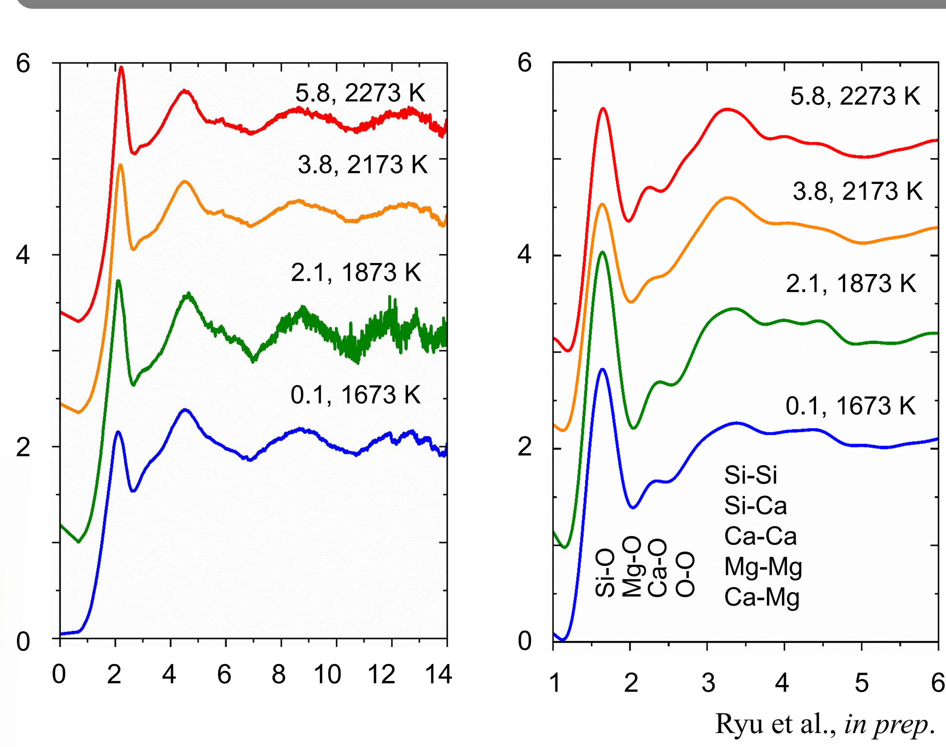
Results and Discussion

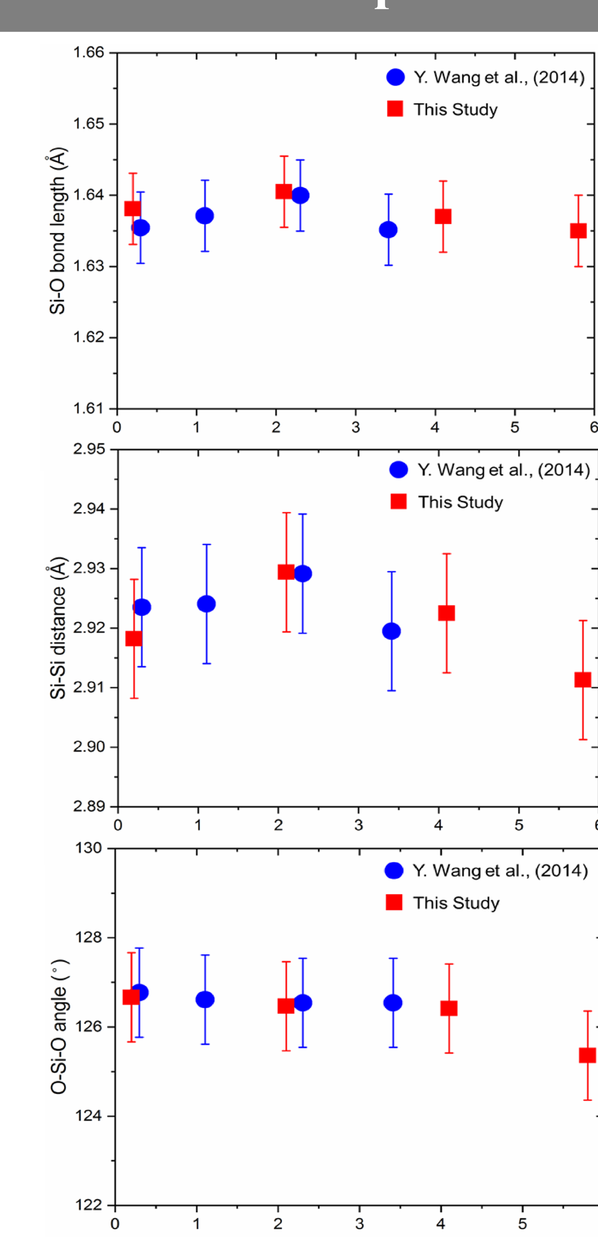
A. Vibrational Modes of Supercooled Liquids



- In-situ confocal solid-state Raman system (GSECARS) was utilized to investigate the pressure-induced vibrational responses of various glass samples.
- The En, Wo, Ab, and Jd were loaded into diamond anvil cell (DAC) to conduct high-pressure and high-temperature experiments.
- Raman spectra obtained from the surface of the recovered samples show typical glass signatures, which can be described by four major groups [7-8]:
  - Localized boson peaks at 0 - 200 cm^{-1}.
  - Metal oxide peaks at 250 – 500 cm^{-1}.
  - Si-O-Si bending and stretching modes at 600 cm^{-1}.
  - Asymmetric and symmetric stretching modes at 850 to 1100 cm^{-1}.
- The frequencies of the Si-O-Si symmetric and asymmetric stretching modes are related to the degree of polymerization of the SiO_4 and AlO_4 tetrahedral network Q^n (n= Q^0, Q^1, Q^2, Q^3 and Q^4).

B. Example of High-Pressure X-ray Diffraction of Diopside Melt

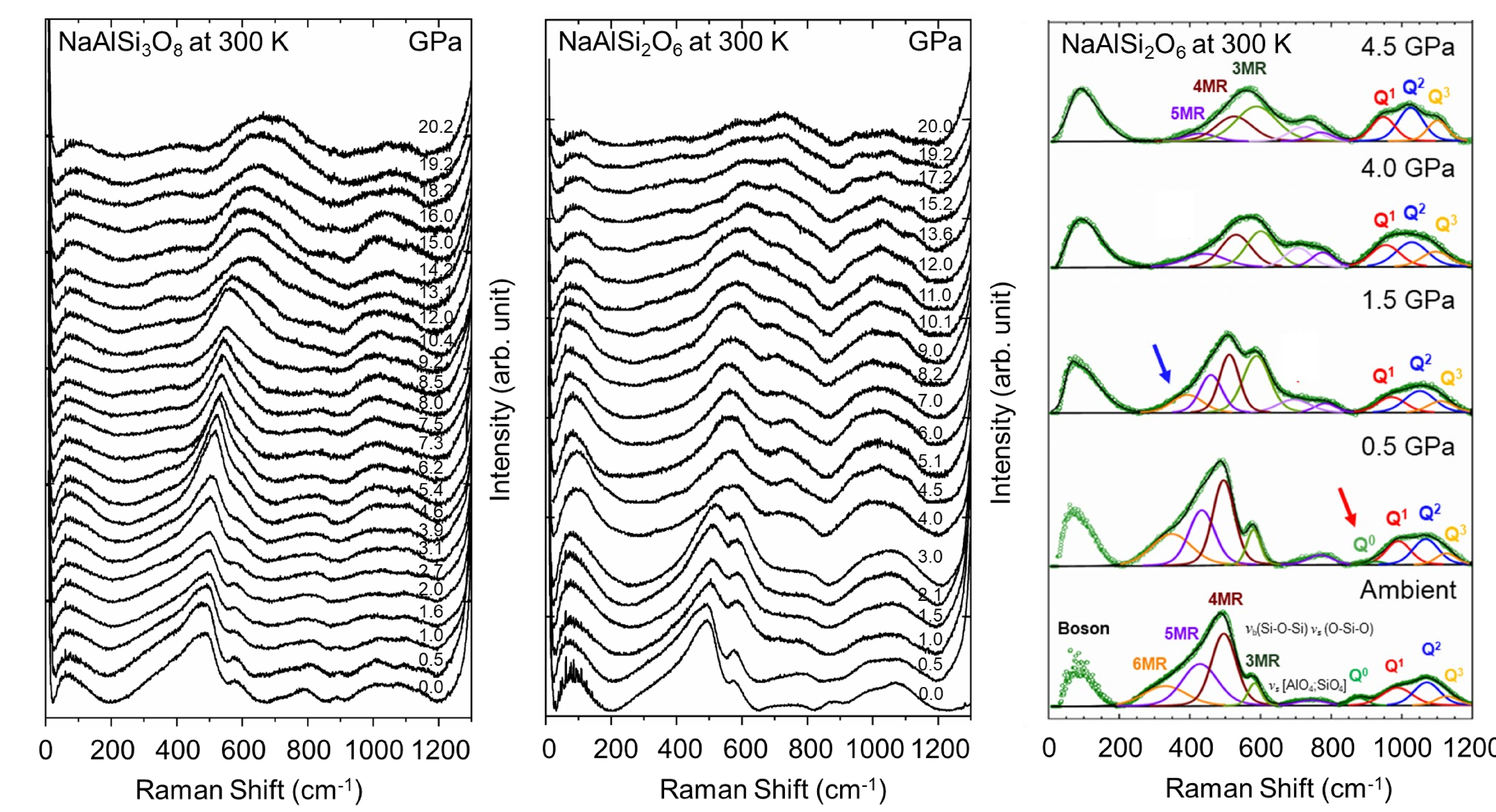




GPa	Temp (K)	Si-O (Å)	Mg-O (Å) ?	Ca-O (Å) ?	Si-Si (Å)	Si-O-Si (° degree)
0.2	1678	1.63(5)	2.07(1)	2.42(1)	2.91(8)	126.6(7)
2.1	1878	1.64(0)	2.08(8)	2.40(2)	2.92(9)	126.4(6)
4.1	2123	1.63(7)	2.09(9)	2.45(9)	2.92(3)	126.4(1)
5.8	2278	1.63(5)	2.07(4)	2.36(7)	2.91(1)	125.3(6)

- Currently, we are working on other silicate melts at HP (above 12 GPa) and HT (~3000 K) to understand structural changes and responses.

C. Raman Spectrum of Albite and Jadeite up to 20 GPa



- Raman spectra of Ab and Jd glasses show the spectral changes associated with the poly-amorphic/chemical transformations.
- Upon initial compression, essentially all Raman modes of Ab and Jd shifted toward high frequencies, except for the defect peak whose intensity decreased continuously.
- The defect peak at ~480 cm^{-1} is characterized by the presence of SiO_4 units in a 3D network.
- Above 0.5 GPa, the Q^0 species disappeared at 870 cm^{-1}. This may indication of the monomer bind with available Q^0 and/or Q^1 species, resulting in an increase in concentration of Q^1 and Q^2 species.
- Above 1.5 GPa, the intensity of the 6MR and 5MR decreases, while the intensity of 3MR increases.
- At ~8 GPa, a new peak around 320 cm^{-1}appeared This peak may indicate higher M-O coordination changes
- Above 12 GPa, the intensity of both the asymmetric and symmetric modes of Ab and Jd decreases. This reduction in intensity might be linked to changes in the selection rules, which are influenced by slight variation in the local ordering of the glass structure as pressure increases [6].

Conclusions

- A Paris-Edinburgh press combined with an MCC has been successfully commissioned at GSECARS Sector 13-IDC for monochromatic X-ray scattering studies on amorphous materials under pressure and high temperature.
- The Raman spectra of Ab and Jd changes with pressure, indicating the presence of structural modifications above ~0.5, 1.5, 8, and 12 GPa.

References

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