

**Project Title:**

**Prediction of Crystal Structure and Properties**

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**Description of the project**

1. Background and purpose of the project, relationship of the project with other projects

Pressure is a very sensitive probe for chemical bonding. Traditionally, much emphasis has been put on breaking new ground in achieving the highest pressure possible in the laboratory using either shock-wave or the diamond anvil techniques. Pressure up to 1 TPa has been achieved. Many new and exotic phenomena, such as quantum liquid phase, superconductivity etc. has been uncovered. Our recent interest is on the properties of minerals and mineral melts at the Earth mantle and Core. Apart from the structures and structural information, the emphasis of this project is understand the transport properties, such as viscosity and thermal conductivity under extreme conditions, such as high pressure and high temperature. For the latter, close to the melting temperature. We wish to answer questions concerning the thermal history of the Earth and also how volcanic activity may be activated due to the transport of the mineral melts. Recently we have implemented a new First Principles Molecular Dynamics method to compute thermal conductivity directly from the atomic trajectory without explicit calculations of the heat flux correlation functions (N.J. English

and J.S. Tse, *Comp. Mat. Sci.*, <http://dx.doi.org/10.1016/j.commatsci.2016>).

Compare the current lattice dynamics based methods employing the Boltzmann Transport Equation approximation, the advantages of this present approach is that anharmonic effects of all orders are automatically included in the MD procedure. The kinetic theory is not invoked in the calculation of the thermal conductivity. Furthermore, systems with impurities can be easily computed.[1]

2. Specific usage status of the system and calculation method

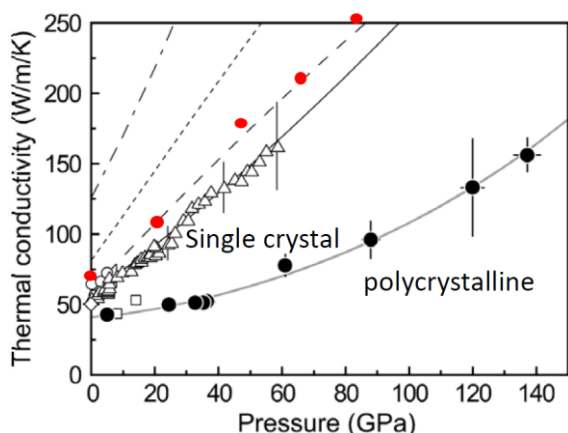
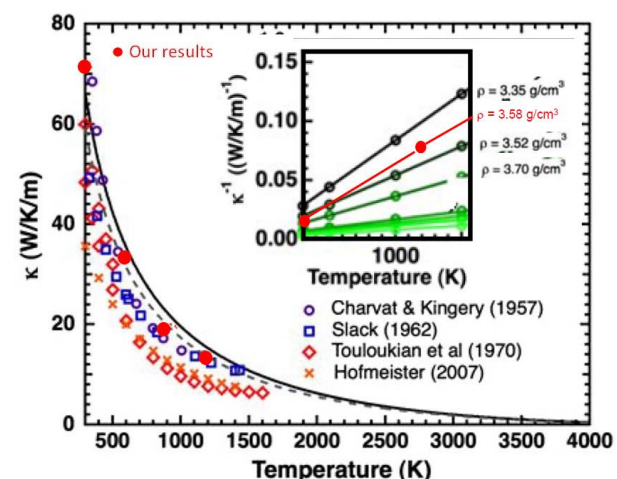
Specific usage status of the system and calculation method

The RIKEN HPCC were used on the validation of the new MD method on the thermal conductivity of pure and doped periclase MgO, a major mineral of the Earth. The computational facility was also used to investigate the structure and transport properties in basalt glasses and melts. In this project, we performed extensive MD calculations from ambient pressure to 80 GPa. From the trajectories, we extracted the structures, equation of state, bulk modulus, velocity of sounds from ambient to 80 GPa.

All calculations were performed using the public domain codes VASP code. Ultrasoft potentials were used for the MgO calculations and PAW + GGA+U was used in the Fe-doped MgO calculations. For the basalt, the PAW potentials were used.

### 3. Result and Conclusion

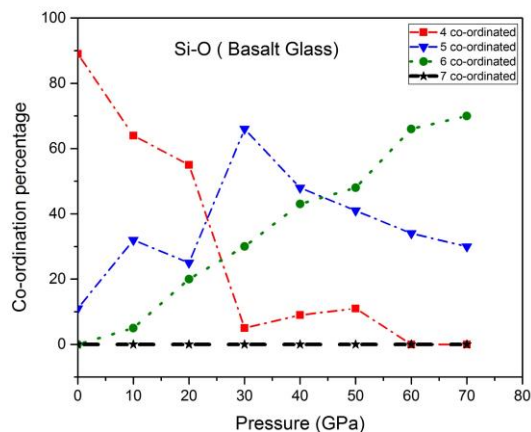
The thermal conductivity of a sample can be computed from the MD trajectory using the Einstein relationship modified for the diffusion of the energy momenta. This method is applicable to all solid phases (crystalline or amorphous). To illustrate the accuracy of the method, the calculated and experimental results under ambient and high pressure (300K) are compared below.



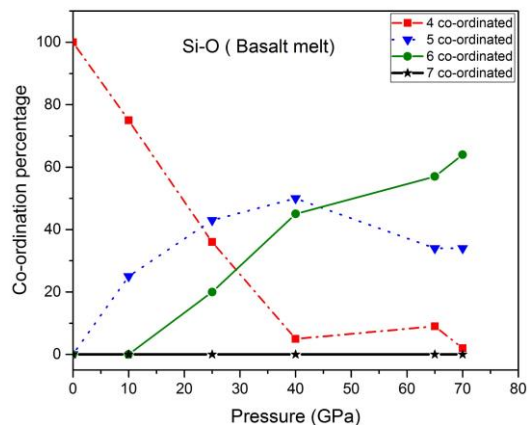
The results show the new implementation is high accurate. Most significantly, we successfully reproduced the surprising observation that Fe doped MgO has a significantly lower thermal conductivity

than the pure materials. Analysis of the latest calculations on Fe-doped MgO, at 300K and 0 GPa, the calculated thermal conductivity of a lightly doped (1.56 mole% Fe) MgO is  $49.3 \pm 4.8$  W/m/K, which is significantly lower than pure MgO of  $73.6 \pm 8.0$  W/m/K. A paper summarizing all the results is now being written and will be submitted for publication in the near future

We illustrate the structural changes from plotting the variation of the Si-O coordination number with applied pressure on basalt glasses and melts below. The most important observation is 4-coordinated Si vanished around 30 GPa in the glass but persisted up to 40 GPa in the melt. The fraction of 5-coordinated Si is higher in the glass than in the melt. This result has important implication on the structural rigidity of the melt which is related the volcanic activity.



## Usage Report for Fiscal Year 2016



#### 4. Schedule and prospect for the future

We wish to complete the Fe-doped periclase thermal conductivities at several experimental Fe dopant concentrations. Direct comparison with experiment will be made. We also need to finish calculations on the basalt melts. The objective is to compute the viscosity which require very long MD simulation times (*ca.* 50 – 100 ps. We are confident that both project will be completed in fiscal 2017

Usage Report for Fiscal Year 2016

**Fiscal Year 2016 List of Publications Resulting from the Use of the supercomputer**

**[Publication]**

Zhi Li, Qiong Liu, Shujuan Han, Toshiaki Iitaka, Haibin Su, Takami Tohyama, Huaidong Jiang, Yongjun Dong, Bin Yang, Fangfang Zhang, Zihua Yang, and Shilie Pan "Nonlinear electronic polarization and optical response in borophosphate BPO<sub>4</sub>", Phys. Rev. B 93, 245125 (2016).

<http://dx.doi.org/10.1103/PhysRevB.93.245125>

Feng Xiaolei, Zhang Jurong, Liu Hanyu, Toshiaki Iitaka, Yin Ketao, Wang Hui "High pressure polyhydrides of molybdenum: A first-principles study", Solid State Communications, Volume 239, p. 14-19. (2016).

<http://dx.doi.org/10.1016/j.ssc.2016.03.025>

Chuanxun Su, Jurong Zhang, Guangtao Liu, Xin Wang, Hui Wang and Yanming Ma, "Catenation of carbon in LaC<sub>2</sub> predicted under high pressure", Phys. Chem. Chem. Phys. 2016, 18, 14286

<http://dx.doi.org/10.1039/C6CP01484D>

**[Oral presentation at an international symposium]**

**J.S. Tse, The 4th Workshop for Extreme Materials Science, RIKEN, Dec. 2016.**

[1] J. S. Tse, *The 4th Workshop for Extreme Materials Science "Thermal Conductivity of Earth"*,

<http://www.iitaka.org/~xmat/event/20161213xmat-workshop4.pdf>