フォルステライト結晶およびガラスの表面構造
Surface Structures of Forsterite Crystal and Glass

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In interstellar molecular clouds, elements such as hydrogen, oxygen, carbon, and nitrogen deposit on dust grains, and form various molecules (e.g., H_2O, CO, CO_2, NH_3, CH_4, H_2CO, CH_3OH, and so on). These molecules undergo chemical evolutions to organic molecules through various processes on the surface of dust grains [1]. Forsterite (Mg_2SiO_4) and enstatite ($MgSiO_3$) have been observed in interstellar molecular clouds and young stellar objects [2]. Although various studies have been performed for bulk structures of forsterite and enstatite, their surface structures are less conclusive [3]. To investigate the surface structures of forsterite in crystalline and glassy states, molecular dynamics (MD) calculations were performed. The surface structure is one of the important factors governing the chemical evolutions in interstellar molecular clouds.
The MD calculations were performed using an atom-atom potential model [4]. The potential parameters were empirically determined by constraining the model to reproduce the experimental results of density, thermal expansion coefficient, and bulk modulus [4]. The glass structure was prepared by quenching the liquid phase consisting of 2400 Mg_2SiO_4 from 3000 K to 10–1750 K with 2 K/fs in rate. An infinite surface was simulated by replicating the cell in the directions parallel to the surface using periodic boundary conditions. The pressure was kept at 0.1 MPa. The MD code was run with NTV ensemble at each temperature for 500 ps with a time step of 0.5 fs. The layer with 0.5 nm in thickness from the outmost atom was analyzed as the surface layer.
The result shows that the melting temperature of forsterite crystal with surface layer was 1927 K. This value is lower than the MD result of bulk state without surface (2418 K [4]) and experimental result (2171 K [5]). This depression of the melting temperature is attributed to the structure and thermal vibrations of atoms in surface layer of forsterite crystal. The nearest Si–Si distance, which was analyzed using the pair correlation functions of atoms, for surface layer of crystal is larger than that of internal part. Furthermore, the amplitudes of thermal vibrations of atoms in surface layer are larger than those of internal part. The results indicate that a surface layer with low density and high thermal vibrations exists in forsterite crystal. The amplitudes of thermal vibrations in surface layer increase with warming and approach the values of the bulk state at its melting point (i.e., 2418 K) at around 1927 K. This induces the depression of the melting temperature for system with the surface layer. For glassy state, a surface layer with short Si–Si distance exists, although the amplitudes of thermal vibrations of atoms are large in comparison with the values of the internal part. This inverted tendency may be resulted from an inhomogeneous structure of surface layer in the glassy state. The surface structures of crystalline and glassy forsterites have important implications for adsorption, diffusion, and chemical reaction in interstellar dust grains.

References:
[1] N. Watanabe, A. Kouchi, *Prog. Surf. Sci.*, **83**, 439 (2008).
[2] J. P. Bradley, L. P. Keller, T. P. Snow, M. S. Hanner, G. J. Flynn, J. C. Gezo, S. J. Clemett, D. E. Brownlee, J. E. Bowey, *Science*, **285**, 1716 (1999).
[3] S. Kohara, K. Suzuya, K. Takeuchi, C. K. Loong, M. Gimsditch, J. K. R. Weber, J. A. Tangeman, T. S. Key, *Science*, **303**, 1649 (2004).

[4] T. Ikeda-Fukazawa, *J. Soc. Inorg. Mater. Jpn.*, **23**, 130 (2016).
[5] B. T. C. Davis, J. L. England, *J. Geophys. Res.*, **69**, 1113 (1964).

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