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Mg-ペロブスカイトにおける3価鉄のスピン転移と下部マントルでの意味

Spin transition of ferric iron in Mg-perovskite and its implication to the lower mantle

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Pressure-induced high spin - low spin transitions of iron in the lower mantle minerals are very important to understand the mineralogy and dynamics of the lower mantle. However, the spin transition of iron in Mg-perovskite, the most abundant mineral in the lower mantle, has been the issue of large controversy. There has been a large discrepancy between the reports of the experimental and theoretical studies, and within the experimental studies the results are inconsistent with each other. We think that these inconsistencies of the experimental results partly originate from the poor quality and/or poor characterization of the measured samples. We measured the spin state of ferric iron in Mg-perovskite up to 200 GPa by X-ray emission spectroscopy (XES) using the well qualified samples, and obtained the results which are almost consistent with the theoretical reports and explain the diversity of the experimental reports.
We measured the spin state of ferric iron in Mg-perovskite with the composition $Mg_{0.85}Fe^{3+}_{0.15}Al_{0.15}Si_{0.85}O_3$ by XES. The obtained XES patterns of ferric iron show the high spin pattern at pressures up to 100 - 120 GPa and gradually change to the low spin pattern at higher pressure, although the XES pattern is still intermediate between the high and low spin patterns even at 200 GPa. These XES patterns at various pressures are well fitted by the linear combination of the high-spin and low-spin patterns of the reference material Fe_2O_3, and the fitted ratio changes gradually, not stepwise, with pressure. These results indicate that the spin state of ferric iron at the A-site of Mg-perovskite is in the high spin state up to 200 GPa, while that at the B-site is in the low spin state from relatively low pressure. The gradual increase of the ratio of the low spin state of ferric iron can be explained by the partial disordering of ferric iron and Al between the A- and B-sites, which would be caused by the annealing (around 1200 - 1500 K) of the samples during the XES measurement. This means that the spin state of ferric iron in the experimental samples of Mg-perovskite will be not simply a state function of pressure and temperature but also affected by the thermal history of the samples.
In the lower mantle, ferric iron- and Al-bearing Mg-perovskite is under high pressure and high temperature, and therefore, the partial disordering of ferric iron and Al between the A- and B-sites would be significant and cause the increase of the ratio of the low spin state of ferric iron in Mg-perovskite.

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