Ultrafast structure transformation of olivine to ringwoodite during laser-driven shock compression

Rocky terrestrial planets, as represented by Earth, were considered to form as consequences of numerous impact events of small planetary bodies 4.6 billion years ago. These events had continued until the completion of planet formation within a dusty disk surrounding the young sun. Although we cannot find any materialistic evidence of these early impact events in the rocks and minerals composing the current Earth, a number of topographical records remaining on surfaces of other smaller bodies in space consistently show that ancient impact events played essential roles in the growth of all solar system bodies. Indeed, many asteroids, for example, have numerous craters left over from ancient impact events, and there is a possibility that the materials in these craters had preserved another type of evidence induced by impact compression. In other words, it is reasonable to expect that the impact events were recorded as not only macroscopic geological evidence such as craters, but also as nanoscopic crystallographic evidence such as rearrangements of mineral structures. The most typical type of evidence of the latter type is the presence of high-density silicate structures within primitive meteorites that were formed in the early solar system and then remained in space until very recently [1].

Olivine [α-$(Mg,Fe)_2SiO_4$] is one of the most popular silicate minerals found on terrestrial planets and other rocky bodies in the solar system. It was previously reported that a fraction of this mineral in some primitive meteorites was transformed into denser structures named wadsleyite [β-$(Mg,Fe)_2SiO_4$] and ringwoodite [γ-$(Mg,Fe)_2SiO_4$] [1]. We have recently discovered that a portion of such natural ringwoodite was once again transformed into another dense structure, ε-$(Mg,Fe)_2SiO_4$, a new mineral that was not observed in the previous research and was named poirierite [2,3]. The macroscopic form of these meteorites containing the dense silicates is often extensively deformed, implying that they had been subjected to shock compression events induced by mutual collisions between their parent asteroids. During these natural shock events, minerals experienced dynamic high-pressure environments where the pressure rapidly increased and decreased with time. We aim to determine these pressure values and their durations induced by such events that triggered the crystallization of the denser minerals. Such work will provide invaluable information for understanding the early evolution history of the solar system. For this purpose, it is necessary to determine the relevant structure transformation mechanism and kinetics that occurred in transient high-pressure environments. Therefore, an ultrafast time-resolving X-ray probe was introduced to analyze the evolution of the crystal structure in laboratory-based shock experiments designed to reproduce the impact events.

The experiments were conducted at SACLA **BL3** EH5, where the formation of a ringwoodite structure within a shock-compressed olivine crystal was observed [4]. Schematic diagrams in Fig. 1 show the experimental setup and the observed structure transformation process. Using a high-power laser system installed onsite at SACLA, a single-crystal sample of synthetic forsterite olivine [α-Mg_2SiO_4] was compressed to reach pressures of 60 to 100 GPa. An optical laser pulse of 532 nm wavelength with a full width at half-maximum of 3.3 ± 0.2 ns was focused into a beam of about 250 μm diameter on the target. A hydrocarbon ablator absorbed this pulse to generate a shock wave that propagated into the olivine crystal to compress its lattice, comprising Mg, Si, and O atoms, along the propagation direction of the wave; this direction was set parallel to the *a*-axis of olivine, as shown in Fig. 1. A femtosecond 10-keV XFEL pulse was focused to be spatially and temporally overlapped with the shock-compressed sample volume. An X-ray diffraction image was immediately obtained to record the crystal structure under shock compression. The experiments were repeated with the variable delay time of the XFEL pulse from the optical laser pulse to analyze the time evolution of the sample structure. Taking into account the crystal axis orientations and the compression direction, as well as the incident angle of the XFEL pulse ω, the time evolution of the specific reflection (such as g = 200 or 300) of olivine was set to be selectively observed as a function of time.

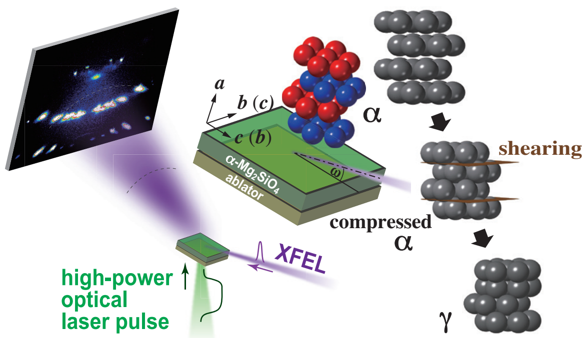

Fig. 1. Experimental setup at SACLA BL3 for analyzing time evolution of forsterite structure under compression, and the observed transformation process.

Figure 2(a) shows the evolution of $g = 200$ as a function of delay time t after the laser pulse arrived on the ablator. Figure 2(b) shows the corresponding one-dimensional diffraction patterns as a function of $d = \lambda/2\sin\theta$. In the olivine structure, oxygen atoms were hexagonally close-packed. The 2θ of the spotty reflections observed in Fig. 2(a) indicates the interlayer distance d_{200}. When the shock wave arrived at olivine at $t = 4$ ns, the position of d_{200} was suddenly shifted to indicate its reduction by shock compression, where it was split into two spots marked with E and P. These were from the regions compressed by the elastic (E) and plastic (P) waves both propagating through the olivine crystal. Both waves decayed with time as they propagated, and the oxygen interlayer distances in these regions were consistently increased. On the other hand, at $t \geq 7$ ns, another reflection marked with D suddenly emerged. It originated from a region of denser structure produced by a third wave that induced the transformation as it propagated. It appeared on the compressed side of P, indicating that the transformed structure had a distinctively higher density than that of the compressed olivine (including both E and P). Its intensity rapidly increased between $t = 6$ ns and 8 ns, indicating that the denser structure crystallized within a duration of a few nanoseconds, where the two cation species, in addition to the oxygen anions, were all appropriately rearranged. Such a complex process has never been observed at this time scale. While olivine has a hexagonal close-packed lattice of oxygen, any of the possible denser structures has a cubic close-packed lattice [2,3]. For this reason, the transformation mechanism must involve the shear deformation of oxygen layers while maintaining their orientation (Fig. 1). Indeed, this is what we observed in Fig. 2(a); it is therefore reasonable to conclude that the ultrafast shear deformation was actually induced. By further analysis of the atomic-scale transformation mechanism from the time evolution of another reflection, $g = 300$ [4,5], it was proved that the product structure was ringwoodite rather than wadsleyite or poirierite.

In summary, an ultrafast transformation mechanism of olivine to ringwoodite through shear deformation is driven efficiently by impact events without any limitation on the minimum body size, as long as the relative velocity between asteroids exceeds the threshold; the transformation proceeds through collisions of meter-sized bodies where the minerals are compressed within microseconds, which is still sufficient to drive the shearing. We suggest that the occurrences of nanoscale platelike dense minerals observed in natural olivine crystals may indicate their having experienced such a collision [4]. Pressure values of 60 to 100 GPa correspond to relative velocities of 5 to 7 km/s, which are the most common in the asteroid belt observed today. If we can extract such a record of the collision history and collision velocity by observing a wide variety of planetary materials, our understanding of the history of material evolution in the solar system will be markedly modified.

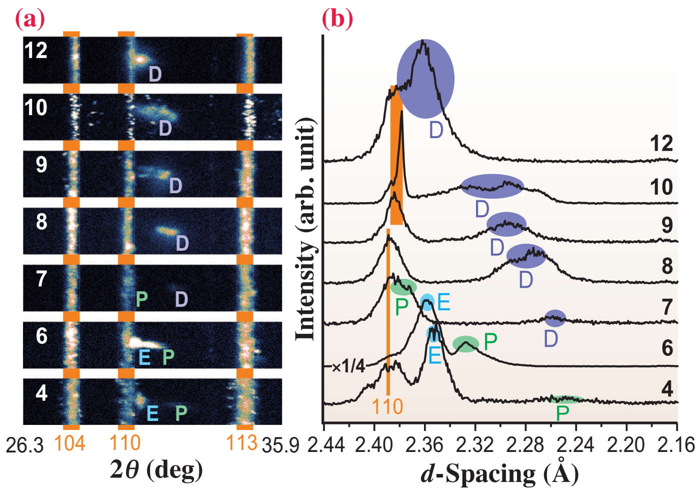

Fig. 2. **(a)** Time evolution of the X-ray diffraction image around the $g = 200$ reflection. The numbers indicate the delay t of the XFEL pulse in nanoseconds where the laser pulse started at $t = 0$. E, P, and D denote reflections originating from the regions compressed by elastic, plastic, and phase transition waves, respectively. In all three regions, the interlayer distance of oxygen stacking layers (see Fig. 1) was compressed parallel to the a-axis direction of forsterite. In these experiments, a thin sheet of polycrystalline Al_2O_3 was attached to the back of the target to indicate the arrival time of the shock wave after it passed through the forsterite. The three orange-indexed lines came from this Al_2O_3 layer. **(b)** X-ray diffraction patterns integrated from (a) as a function of $d = \lambda/2sin\theta$, where λ is the wavelength (1.24 Å) and θ is the diffraction angle. Note that d decreases to the right.

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