ELSEVIER

Earth and Planetary Science Letters 147 (1997) E9–E15

EPSL

Express Letter

Single-crystal elasticity of β-Mg_2SiO_4 to the pressure of the 410 km seismic discontinuity in the Earth's mantle

Chang-sheng Zha [a,*], Thomas S. Duffy [b], Ho-kwang Mao [a], Robert T. Downs [c], Russell J. Hemley [a], Donald J. Weidner [d]

[a] *Geophysical Laboratory and Center for High-Pressure Research, Carnegie Institution of Washington, 5251 Broad Branch Road NW, Washington, DC 20015, USA*

[b] *Department of Geosciences, Princeton University, Princeton, NJ 08544, USA*

[c] *Department of Geological Sciences, University of Arizona, Tucson, AZ 85721, USA*

[d] *Department of Earth and Space Sciences and Center for High-Pressure Research, State University of New York, Stony Brook, NY 11794, USA*

Received 2 December 1996; revised 7 January 1997; accepted 9 January 1997

Abstract

The complete set of elastic stiffness coefficients of a single crystal of Mg_2SiO_4 in the modified spinel (wadsleyite) structure are reported at eight pressures from ambient to 14 Gigapascals (GPa). All moduli vary linearly with pressure within the resolution of the data. Third-order Birch–Murnaghan equation fits to aggregate elastic moduli yield values of 170 ($\pm$2) GPa and 4.3 ($\pm$0.2) for the adiabatic bulk modulus and its first pressure derivative, and 115 ($\pm$2) GPa and 1.4 ($\pm$0.2) for the shear modulus and its first pressure derivative. These data, together with previous results for α-Mg_2SiO_4, enable us to make the first in situ determination of the velocity increase due to the α–β transition at the pressure ($\sim$ 13.8 GPa) of the 410 km discontinuity. The room-temperature velocity increase at the transition pressure is 9.8% for compressional waves and 12.4% for shear waves.

Keywords: transition zones; elasticity; spinel; upper mantle

1. Introduction

The transition zone of the Earth's mantle, defined by seismic discontinuities near 410 and 670 km depth, has long been recognized as one of the most important and enigmatic regions of the Earth's interior [1]. A detailed knowledge of the phase and chemical changes responsible for the seismic structure of Earth's upper mantle is essential for achieving an integrated theory that can describe the evolution, dynamics, and physical state of the mantle. Of critical importance for understanding seismic observations is the change in elastic properties across the transformation sequence in $(Mg,Fe)_2SiO_4$ from the olivine (α) to modified spinel (β) to spinel (γ) structure.

Upper mantle mineralogical models satisfying one-dimensional seismic velocity profiles can be constructed from mineral elasticity and phase equilibria data. The previous generation of upper mantle models [2,3] were unable to reach a consensus regarding upper mantle mineralogy due to the lack of

* Corresponding author. E-mail: zha@granite.ciw.edu

PII S0012-821X(97)00010-1

direct elasticity measurements at high pressure and temperature. This has motivated considerable efforts to extend mineral sound velocity measurements to higher pressures [4–10]. While the single-crystal elastic properties of the olivine phase are now reasonably well characterized at high pressure [5,7–9], there have previously been no such measurements for the high-pressure polymorphs.

2. Experimental method

Single-crystal samples of β-Mg_2SiO_4 were synthesized in a large volume press at 18.8 GPa and 1500°C. Two high quality samples were selected and polished to flat, parallel plates with thicknesses of 30 and 50 μm. Each was polished within a few degrees of the crystallographic plane which intersects the three crystal axes at equal angles. Electron microprobe analysis demonstrated that the samples were pure Mg_2SiO_4. Electron microscopy showed that submicron inclusions are present in restricted areas for both samples. Raman spectroscopy revealed no peaks that could be attributed to hydroxyl. Single-crystal X-ray diffraction was performed at ambient and at selected elevated pressures (1.8 GPa, 3.2 GPa and 10.2 GPa). The ambient pressure volume was 535.8 ($\pm$0.2) Å^3, consistent with a previous determination [11]. High-pressure volumes also agree with the earlier data, as well as with the equation of state from Brillouin scattering determined here.

Acoustic velocities were measured using Brillouin scattering in a diamond anvil cell. There are a number of potential problems that can arise when making such measurements, including restricted optical access and overlap of Brillouin peaks from the pressure medium and the sample. We have recently succeeded in developing techniques that overcome these problems and allow for determination of the complete set of elastic stiffness coefficients at high pressures for materials of orthorhombic and hexagonal symmetries [6–8]. However, when examining materials with high acoustic velocities, such as high-pressure polymorphs, compressional Brillouin peaks from the sample fall within the same spectral region as the shear peaks from the diamond anvils. Since the volume of diamond is much larger than the volume of sample material, the diamond Brillouin peaks completely obscure the sample peaks. Since the two diamonds will also, in general, have different crystal orientations, each will contribute its own Brillouin peaks and a large spectral region can be affected. For β-Mg_2SiO_4, the compressional wave velocities of the sample were obscured by the diamond shear wave peaks at pressures above 10 GPa. To overcome this problem, a long focal length cylindrical lens together with a spatial filter were used to reduce the astigmatism introduced by the anvils. This method was found to reduce the intensity of the diamond signal by more than 90% for a 30 μm thick sample (Fig. 1). The astigmatism correction also markedly reduces the size of the peaks from the pressure medium. While it was not possible to remove the diamond signal completely, it was sufficiently de-

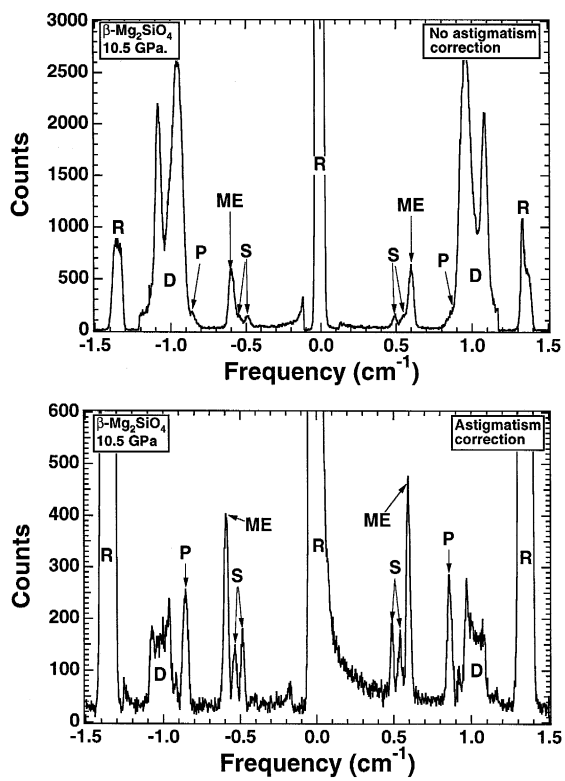

Fig. 1. Comparison of Brillouin spectra of wadsleyite at 10.5 GPa before (upper panel) and after (lower panel) astigmatism correction. Peaks: R = unshifted Rayleigh peaks; P and S = compressional and shear peaks in β-Mg_2SiO_4, respectively; ME = peaks from the methanol–ethanol pressure medium; D = peaks from the diamond anvils. Similar data collection times were used in obtaining both spectra.

pressed that the sample compressional wave signal could be readily observed.

The 50 μm thick sample was loaded into a diamond anvil cell together with an argon pressure transmitting medium and a grain of ruby for pressure calibration. Brillouin spectra were recorded at 3.1, 6.3, 8.1, and 9.4 GPa. After pressure release, the sample was recovered and Brillouin spectra were measured at ambient pressure. The 30 μm thick sample was loaded in a separate diamond cell with a 4:1 methanol–ethanol mixture as a pressure medium. Brillouin spectra were recorded at 10.1 and 10.5 GPa. This sample was then unloaded, recovered, and reloaded using a helium pressure medium. Brillouin spectra were recorded at 14.2 GPa. However, premature failure of the diamond anvils resulted in loss of this sample during pressure release from 14.2 GPa.

3. Results

At each pressure, the compressional and two shear acoustic velocities were determined at about 18 directions at 10° intervals within the plane perpendicular to the diamond cell axis. A total of 400 acoustic velocities, representing more than 400 h of total data collection time, were recorded in this study. The data were inverted using non-linear least-squares to obtain the nine independent elastic moduli as a function of pressure (Fig. 2, Table 1). Previous ambient pressure data [12] are also shown in the figure; this is the first determination of individual elastic moduli at high pressure for this material. All elastic moduli vary linearly with pressure within the resolution of the data.

Bounds on the bulk and shear moduli of a randomly oriented polycrystalline aggregate of β-Mg_2SiO_4 were obtained from the single-crystal moduli using Voigt–Reuss–Hill averages [13] (Fig. 3). The Voigt and Reuss limits differ by 0.3–0.9% for the bulk modulus and by 1.2–2.8% for the shear modulus. Fitting the data to the Birch–Murnaghan equation yields a value of 170 ($\pm$2) GPa for the bulk modulus, K_{0S}, and 4.3 ($\pm$0.2) for its pressure derivative, K'_{0S}. Values of 115 ($\pm$2) GPa and 1.4 ($\pm$0.2) are obtained for the shear modulus, G_0, and its pressure derivative, G'_0. For the ambient pressure moduli, these values are in reasonable agreement with previous single-crystal elasticity data [12] for this phase (K_{0S} = 174 GPa, G_0 = 114 GPa). Aggregate sound velocity measurements were previously reported for a slightly porous, polycrystalline aggregate of β-Mg_2SiO_4 to 3 GPa using ultrasonic techniques [9]. The pressure derivatives obtained in that study [K'_0 = 4.8 ($\pm$0.1) and G'_0 = 1.8 ($\pm$0.1)] are higher than those found here. More recently, these measurements were extended to 12 GPa using a large-volume press [10]. When fit in the same manner as our data (i.e., to third-order finite strain equations), the pressure derivatives (K'_0 = 4.5, G'_0 =

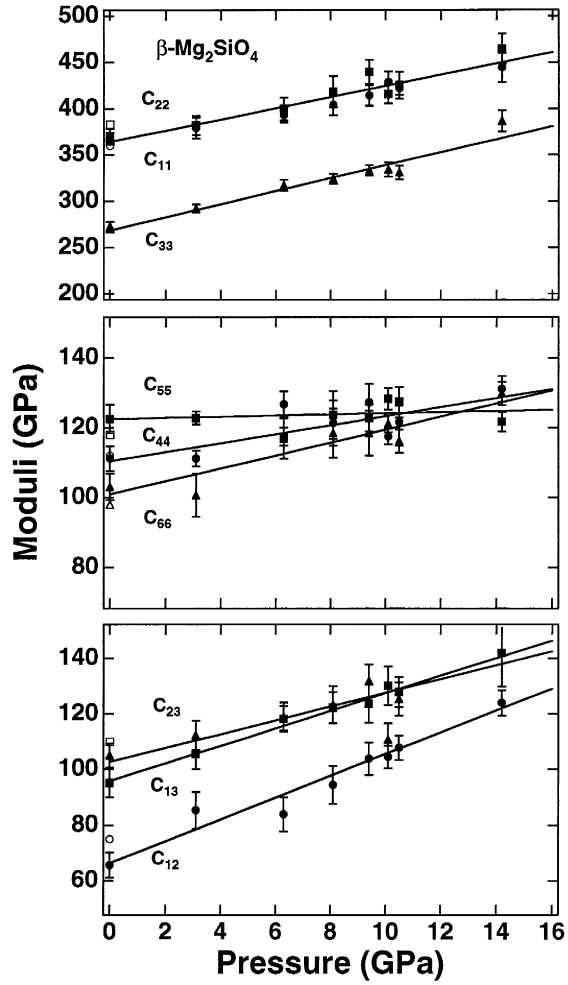

Fig. 2. Single-crystal elastic moduli of wadsleyite as a function of pressure. Filled symbols are the present data with 2σ uncertainties. The solid lines are linear fits to the data. A single line is used to fit the data for C_{11} and C_{22}. Previous ambient pressure data [12] are shown as open symbols.

Table 1
Elastic moduli of β-Mg_2SiO_4

P	C_{11}	C_{22}	C_{33}	C_{44}	C_{55}	C_{66}	C_{12}	C_{13}	C_{23}
0	370.5	367.7	272.4	111.2	122.5	103.1	65.6	95.2	105.1
	7.8	6.5	5.8	3.6	4.0	3.9	4.5	5.2	4.4
3.1	379.3	382.0	292.2	111.2	122.7	100.7	85.4	105.5	112.4
	11.5	10.4	4.5	2.2	1.8	6.1	6.6	5.4	5.3
6.3	393.4	399.9	316.7	126.6	117.5	117.0	83.9	118.2	119.1
	7.9	12.3	6.1	3.7	2.6	5.8	6.1	4.6	5.0
8.1	404.2	418.3	324.0	121.4	123.6	118.5	94.4	122.3	123.4
	11.2	16.8	5.4	6.4	6.7	7.0	6.8	5.5	6.7
9.4	414.4	439.8	333.1	127.1	122.8	118.4	103.9	123.8	131.8
	11.2	13.0	5.8	5.3	3.8	6.4	5.9	6.8	5.9
10.1	428.8	416.0	333.9	117.6	128.2	121.1	104.5	130.2	110.8
	11.3	9.9	7.9	2.2	3.0	2.4	4.1	6.9	5.8
10.5	421.9	425.7	330.7	121.7	127.2	116.2	107.8	127.9	125.4
	6.7	14.2	7.7	6.8	4.3	3.3	4.4	5.4	6.0
14.2	444.5	464.9	386.8	130.9	121.7	129.7	123.9	142.0	151.9
	15.9	16.1	11.3	3.7	2.8	3.3	4.6	12.2	19.6

All quantities are in GPa. Uncertainties (2σ) are listed below each value.

1.6) are intermediate between our results and the previous ultrasonic studies. The use of polycrystalline samples, which retain some porosity even at the highest pressures, may be responsible for these differences.

4. Discussion

One of the advantages of single-crystal elasticity measurements is that they define the complete velocity surface at high pressure. This is particularly

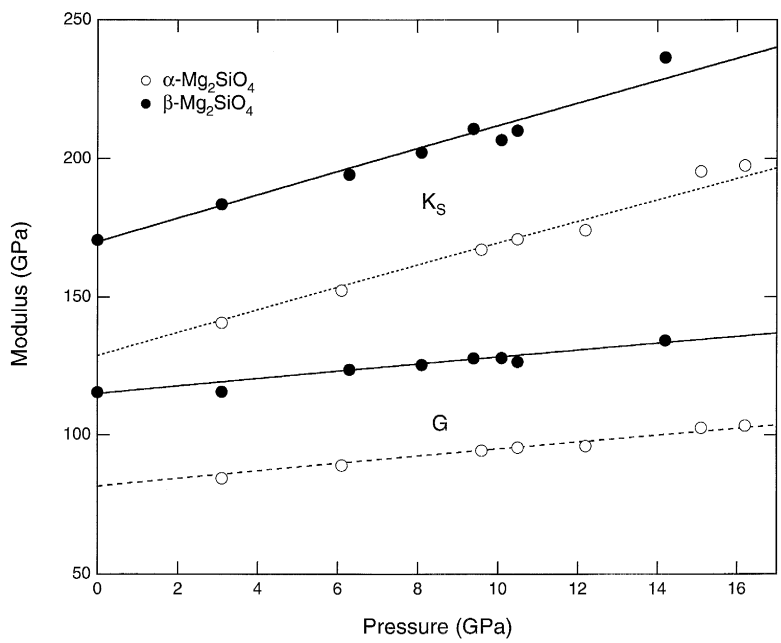

Fig. 3. Bulk (KS) and Shear (G) moduli of wadsleyite and forsterite [7,8] as a function of pressure. Symbols are Voigt–Reuss–Hill averages and solid lines are third-order finite strain fits to the data. Uncertainties in the VRH averages are approximately the size of the symbols.

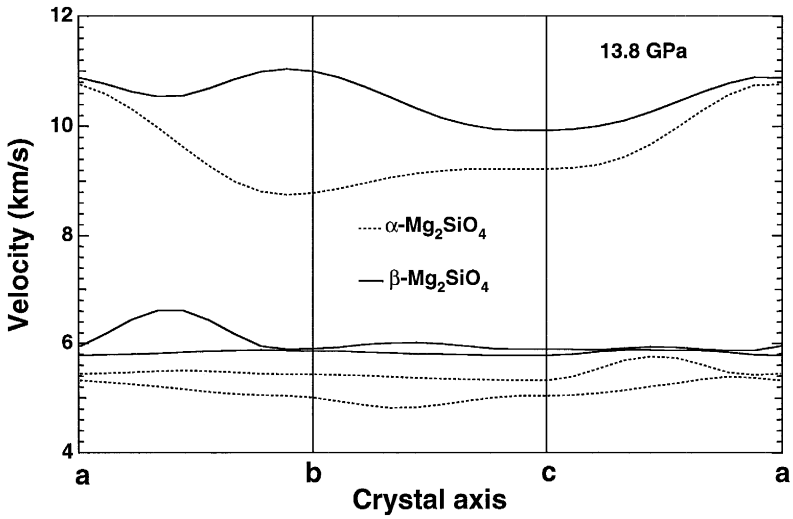

Fig. 4. Sound velocity distributions in the α and β forms of Mg_2SiO_4 at 13.8 GPa. The data are shown for the three principal planes, *ab*, *bc*, and *ca*. Forsterite data are from [7,8]. Linear least-squares fits to the individual moduli (e.g., Fig. 2) were used to compute the moduli at 13.8 GPa.

important for understanding the seismic anisotropy of the Earth's upper mantle [14–16]. Fig. 4 shows a comparison of the velocity distributions in the principal planes for α- and β-Mg_2SiO_4 at 13.8 GPa, approximately the pressure of the 410 km discontinuity in the mantle. Near the *a* direction and throughout most of the *a*–*c* plane, compressional sound velocities in forsterite and wadsleyite are similar. For

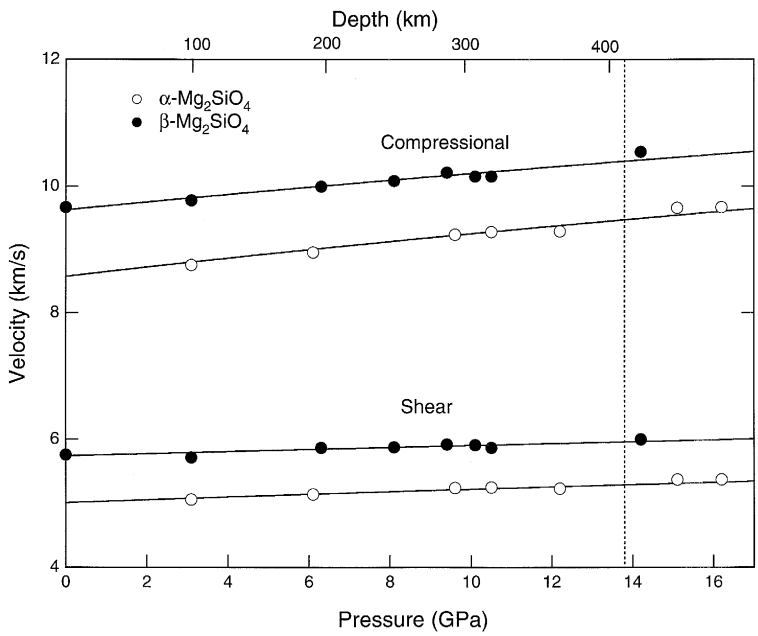

Fig. 5. Aggregate compressional and shear wave velocities of wadsleyite and forsterite from single-crystal Brillouin scattering measurements [7,8] as a function of pressure and depth. ○ and ● = experimental data. Solid lines are calculated using the third-order Eulerian finite strain equations. The vertical dashed line corresponds to the 410 km discontinuity.

compressional velocities, the major difference between the two phases is along the *b* direction. This is the minimum velocity direction in forsterite, but the maximum velocity direction in β-Mg_2SiO_4. Within the principal planes, neither form of the material exhibits a large amount of anisotropy for shear velocities. The velocity anisotropy is a weakly decreasing function of pressure. The compressional velocity anisotropy for β-Mg_2SiO_4 is 17% at ambient pressure, but is in the range of 10–14% at pressures between 10 and 14 GPa. Similarly, the maximum shear wave birefringence is 18% at ambient pressure, but is found to be 13–14% at the highest pressures.

The present results, together with previously reported data for forsterite to 16 GPa [7,8], allow us to make the first direct determination of the velocity difference between the α and β phases of Mg_2SiO_4 at the pressure of the 410 km discontinuity. Compressional and shear velocities for randomly oriented polycrystalline aggregates for both phases are shown in Fig. 5. At ambient pressure, the velocity contrast between the two phases is 12.3% for compressional waves and 14.2% for shear waves. At 13.8 GPa, these differences have been reduced to 9.8% for compressional waves and 12.4% for shear waves. Typical seismic velocity contrasts across the 410 km discontinuity are in the range of 4–5% [17–19]. These are 31–51% of the value of the room-temperature α–β velocity jump at this depth. In an earlier analysis [7], which used extrapolated low-pressure data [4] to characterize the elasticity of β-Mg_2SiO_4, it was concluded that, based on the magnitude of the 410 km discontinuity, the olivine faction of the mantle at this depth is less than 40%. When the present results for wadsleyite are substituted into this analysis, the maximum allowable olivine fraction of the mantle increases to 50% by volume [20]. Future refinement of these values can be obtained from measurements of the effect of temperature on the velocity contrast across the discontinuity.

Acknowledgements

This research was supported by the NSF. The Center for High-Pressure Research is an NSF Science and Technology Center. ***[FA]***

References

[1] F. Birch, Elasticity and constitution of the Earth's Interior, J. Geophys. Res. 57, 227–286, 1952.
[2] T.S. Duffy and D.L. Anderson, Seismic velocities in mantle minerals and the mineralogy of the upper mantle, J. Geophys. Res. 94, 1895–1912, 1989.
[3] D.J. Weidner and E. Ito, Mineral physics constraints on a uniform mantle composition, in: High-Pressure Research in Mineral Physics, M.H. Manghnani and Y. Syono, eds., pp. 439–446, Terrapub, Tokyo, 1987.
[4] G.D. Gwanmesia, S. Rigden, I. Jackson and R.C. Liebermann, Pressure dependence of elastic wave velocity for β-Mg_2SiO_4, and the composition of the Earth's mantle, Science 250, 794–797, 1990.
[5] J.M. Zaug, E.H. Abramson, J.M. Brown and L.J. Slutsky, Sound velocities in olivine at earth mantle pressures, Science 260, 1487–1489, 1993.
[6] C.S. Zha, T.S. Duffy, H.K. Mao and R.J. Hemley, Elasticity of hydrogen to 24 GPa from single-crystal Brillouin scattering and synchrotron x-ray diffraction, Phys. Rev. B 48, 9246–9255, 1993.
[7] T.S. Duffy, C.S. Zha, R.T. Downs, H.K. Mao and R.J. Hemley, Elasticity of forsterite to 16 GPa and the composition of the upper mantle, Nature 378, 170–173, 1995.
[8] C.S. Zha, T.S. Duffy, R.T. Downs, H.K. Mao and R.J. Hemley, Sound velocity and elasticity of single-crystal forsterite to 16 GPa, J. Geophys. Res. 101, 17,535–17,545, 1996.
[9] G. Chen, B. Li and R.C. Liebermann, Selected elastic moduli of single-crystal olivines from ultrasonic experiments to mantle pressures, Science 272, 979–980, 1996.
[10] B. Li, G.D. Gwanmesia and R.C. Liebermann, Sound velocities of olivine and beta polymorphs of Mg_2SiO_4 at Earth's transition zone pressures, Geophys. Res. Lett. 23, 2259–2262, 1996.
[11] R.M. Hazen, J. Zhang and J. Ko, Effects of Fe/Mg on the compressibility of synthetic wadsleyite: β-$(Mg_{1-x}Fe_x)_2SiO_4$ ($x \leq 0.25$), Phys. Chem. Mineral. 17, 416–419, 1990.
[12] H. Sawamoto, D.J. Weidner, S. Sasaki and M. Kumazawa, Single-crystal elastic properties of the modified spinel (beta) phase of magnesium orthosilicate, Science 224, 749–751, 1984.
[13] J.P. Watt and L. Peselnick, Clarification of the Hashin–Shtrikman bounds on the effective elastic moduli of polycrystals with hexagonal, trigonal, and tetragonal symmetries, J. Appl. Phys. 51, 1525–1531, 1980.
[14] P. Silver and W.W. Chan, Implications for continental structure and evolution from seismic anisotropy, Nature 335, 34–39, 1988.
[15] T. Tanimoto and D.L. Anderson, Mapping convection in the mantle, Geophys. Res. Lett. 11, 287–290, 1984.
[16] S.J. Kaneshima, Seismic anisotropy: A review of studies by Japanese researchers, J. Phys. Earth 43, 301–320, 1995.
[17] G. Nolet, S.P. Grand and B.L.N. Kennett, Seismic heterogeneity in the upper mantle, J. Geophys. Res. 99, 23,753–23,766, 1994.

[18] S.P. Grand and D.V. Helmberger, Upper mantle shear structure of North America, Geophys. J.R. Astron. Soc. 76, 399–438, 1984.

[19] J. Mechie, A.V. Egorkin, K. Fuchs, T. Ryberg, L. Solodilov and F. Wenzel, P-wave mantle velocity structure beneath northern Eurasia from long-range recordings along the profile Quartz, Phys. Earth Planet. Inter. 79, 269–286, 1993.

[20] C.S. Zha, T.S. Duffy, R.T. Downs, H.K. Mao, R.J. Hemley and D.J. Weidner, Single-crystal elasticity of the α and β polymorphs of Mg_2SiO_4 at high pressure, in: High-Pressure Temperature Research: Properties of Earth and Planetary Materials, M.H. Manghnani and T. Yagi, eds., in press, 1997.