

Sound velocities and elasticity of aluminous MgSiO₃ perovskite: Implications for aluminum heterogeneity in Earth's lower mantle

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Received 8 March 2004; revised 12 April 2004; accepted 19 April 2004; published 27 May 2004.

[1] Aluminum has been reported to have a remarkably strong effect on the thermoelastic properties of MgSiO₃ perovskite. However, the sound velocities of aluminous MgSiO₃ perovskite have not been previously measured, even though this phase likely dominates most of the chemistry in Earth's lower mantle. Here we report the first sound velocity measurements on aluminous MgSiO₃ perovskite using Brillouin spectroscopy and obtain the following values for the room-pressure room-temperature adiabatic bulk and shear moduli: $K_S = 252 \pm 5$ GPa and $\mu = 165 \pm 2$ GPa, respectively. The presence of 5.1 ± 0.2 wt.% Al₂O₃ in MgSiO₃ perovskite decreases the shear modulus by 5.6%. However, within experimental uncertainties, there is no discernable effect of aluminum on the bulk modulus. We find that variations in the aluminum content of MgSiO₃ perovskite may provide an explanation for some observed lateral heterogeneity in Earth's lower mantle. **INDEX TERMS:** 3909 Mineral Physics: Elasticity and anelasticity; 3919 Mineral Physics: Equations of state; 5102 Physical Properties of Rocks: Acoustic properties; 8124 Tectonophysics: Earth's interior—composition and state (1212). **Citation:** Jackson, J. M., J. Zhang, and J. D. Bass (2004), Sound velocities and elasticity of aluminous MgSiO₃ perovskite: Implications for aluminum heterogeneity in Earth's lower mantle, *Geophys. Res. Lett.*, 31, L10614, doi:10.1029/2004GL019918.

1. Introduction

[2] The chemical composition of Earth's lower mantle (670–2900 km depth, representing roughly 60% of Earth's volume) is controversial, but a point of broad agreement is that it is likely dominated by a perovskite-structured phase in which MgSiO₃ is the main chemical component [e.g., Knittle and Jeanloz, 1987; Irifune, 1994; Fiquet et al., 2000; Andraut et al., 2001]. This perovskite also contains most of the aluminum [Irifune, 1994] (likely in the range of 2–8 wt.% [Bass and Anderson, 1984; Ringwood, 1991]) and much of the iron in the lower mantle. The best resolved properties throughout this region are seismologically-determined sound velocities, which directly probe the in-situ state of mantle material with high spatial resolution. Therefore,

accurate determination of the sound velocities of aluminum-bearing silicate perovskite is essential for understanding the chemical and thermal state of Earth's lower mantle. Recent experiments and density functional theory (DFT) calculations have suggested that such amounts of aluminum dissolved in magnesium silicate perovskite drastically affect the bulk modulus and other thermoelastic properties of silicate perovskite [Zhang and Weidner, 1999; Brodholt, 2000; Kubo et al., 2000; Andraut et al., 2001; Daniel et al., 2001]. Most of the previous experimental studies of silicate perovskite elasticity have been limited to static compression experiments, which yield the isothermal bulk modulus (K_{0T}) through the relation: $K_{0T} = -V(dP/dV)_T$, where V is the volume and P is the pressure. Although x-ray diffraction results provide valuable constraints on the equation of state (EOS), they do not provide information on the shear elastic modulus (μ). Moreover, the few measurements carried out to date on the P-V relationship of aluminous MgSiO₃ perovskite (hereafter referred to as Al-Pv) have yielded contradictory results for K_{0T} . Some studies report that K_{0T} for Al-Pv is $\sim 10\%$ lower than that of MgSiO₃ perovskite (Mg-Pv) [Zhang and Weidner, 1999; Kubo et al., 2000; Daniel et al., 2001], whereas a recent study implied that K_{0T} for Al-Pv is higher than that of Mg-Pv [Andraut et al., 2001]. These discrepancies seriously limit our ability to constrain lower mantle chemistry. Scattering techniques that are direct probes of acoustic phonons, such as Brillouin spectroscopy, provide a means of determining the bulk modulus that is entirely independent of the static compression methods used thus far. In addition, Brillouin spectroscopy provides information on 1) the shear modulus, which cannot be obtained from hydrostatic compression experiments, and 2) sound velocities, which are more directly comparable with observed seismic wave velocities in Earth's interior. In this paper, we present the first measurements of sound velocities of aluminous magnesium silicate perovskite.

2. Experiment

2.1. Sample Characterization

[3] The Al-Pv sample used in the Brillouin experiments was synthesized from a synthetic glass containing 80 mol% MgSiO₃ and 20 mol% Mg₃Al₂Si₃O₁₂ at ~ 25 GPa and 1873 K for 2 hours using a 2000-ton Kawai-type multi-anvil

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Table 1. Isotropic Aggregate Properties for Aluminous and Pure MgSiO_3 Perovskite from Recent Experiments

Studies	V_0 (\AA^3)	ρ (g/cm^3)	V_S (km/s)	V_P (km/s)	μ (GPa)	K_S (GPa)
MgSiO_3 Pv (sc) ^a	162.40(5)	4.106(1)	6.53(3) ^f	10.88(6) ^f	175(2) ^f	253(3) ^f
MgSiO_3 Pv (poly) ^a	162.40(5)	4.106(1)	6.47(6) ^f	10.84(2) ^f	172(3) ^f	253(5) ^f
MgSiO_3 Pv (poly) ^b	162.3	4.06(2)	6.51	-	176(5)	-
MgSiO_3 Pv (pwd) ^c	162.27(1)	4.11	-	-	-	262(9) ^c
Al-Pv (poly) ^d	163.18(8)	4.078(2)	-	-	-	237(2) ^d
Al-Pv (poly) ^e	163.26(1)	4.081(2)	6.35(5)	10.75(5)	165(2)	252(5)

Uncertainties are given in parentheses for the last reported significant digit.

Notes: sc – single crystal, poly – polycrystal, pwd – powdered.

^a*Sinogeikin et al.* [2004].

^b*Sinelnikov et al.* [1998].

^cFrom *Fiquet et al.* [2000, optimized model]: K_S was calculated from $K_S = K_{0T}(1 + \alpha\gamma T)$ using $K_{0T} = 259.5$ GPa, $\alpha = 2.21 \times 10^{-5} \text{ K}^{-1}$, and $\gamma = 1.4$ at 300 K.

^dFrom *Zhang and Weidner* [1999]: MgSiO_3 perovskite with ~ 5 wt.% Al_2O_3 . K_S was calculated using $K_{0T} = 234$, $\alpha = 2.73 \times 10^{-5} \text{ K}^{-1}$, and $\gamma = 1.4$ (as in [c]) at 300 K.

^eThis study. MgSiO_3 perovskite with 5.1(2) wt.% Al_2O_3 synthesized under similar conditions as the sample used in [d]. The density was calculated from the measured x-ray volume and EMP analyses.

^fVoigt-Reuss-Hill average.

apparatus. An angle-dispersive x-ray diffraction pattern of the sample showed all of the expected diffraction peaks of perovskite (space group $Pbnm$). In addition, we observe a single broad, weak diffraction peak at $d = 2.965 \text{ \AA}$ which cannot be indexed by perovskite. Synchrotron x-ray diffraction work [I. Daniel et al., Effect of aluminum on the compressibility of silicate perovskite, submitted to *Geophysical Research Letters*, 2004, hereinafter referred to as Daniel et al., submitted manuscript, 2004] on a sample from the same run charge as that which was used in this study indicates that this peak may correspond to the most intense peak of stishovite, indicating a trace ($\leq 1\%$) amount of stishovite in the sample. However, the effect on our Brillouin results would be much less than our cited uncertainties.

[4] The density of Al-Pv was obtained from the unit-cell volume determined by x-ray diffraction (Table 1) and microprobe (EMP) chemical analyses. For EMP analysis, the focused electron beam conditions were 10 nA incident current and 15 keV. An enstatite glass containing 5 wt.% Al_2O_3 was used as the standard. The results of 15 EMP analyses on Al-Pv give the following oxide weight percents, with standard deviations reported as uncertainties: $\text{SiO}_2 = 57.8 \pm 1.1$, $\text{MgO} = 38.6 \pm 0.6$, $\text{Al}_2\text{O}_3 = 5.1 \pm 0.2$, and $\Sigma = 101.5 \pm 1.3$. The presence of oxygen vacancies may be inferred from the oxide sum exceeding 100%. However, if oxygen vacancies form due to the substitution of Al on Si sites only [e.g., *Brodholdt*, 2000], then a Mg/Si ratio greater than unity would result. In our sample, $\text{Mg/Si} = 0.99 \pm 0.01$, and the sum of cations equals 2 when calculated on the basis of 3 oxygen atoms. This indicates that the sample is not likely to be oxygen deficient. An energy dispersive spectroscopic scan showed that Si, Mg, and Al are the only cations present in this sample. A scan over $50 \mu\text{m}$ at $0.5 \mu\text{m}$ intervals for Si, Mg, and Al indicated that the sample is homogeneous. The average grain size was determined to be $2 \mu\text{m}$.

[5] Optical examination of the sample under cross-polarized light showed random extinction of the individual crystals, indicating the lack of any preferred orientation. We further tested the sample for preferred orientation with x-ray diffraction. A portion of the polycrystalline sample used for Brillouin scattering measurements was mounted on a Bruker Platform 3-circle goniometer equipped with a SMART CCD detector and a crystal-to-detector distance of 4.99 cm. A diffraction pattern was taken using mono-

chromatic $\text{MoK}\alpha$ radiation and a 300 second exposure. The sample was not rotated in the beam to produce the least favorable conditions for obtaining uniform diffraction intensity and any preferred orientation should be obvious. The resulting x-ray image (Figure 1) is grainy, due to the finite number of crystallites, but shows highly uniform intensity around the diffraction ring. This suggests that there is no texture in the sample.

2.2. Brillouin Scattering Measurements

[6] A portion of the polycrystalline Al-Pv sample was polished to a thickness of $40 \mu\text{m}$ (with lateral dimensions of $500 \times 330 \mu\text{m}^2$) for Brillouin measurements at ambient conditions. Brillouin scattering was performed in a 90-degree symmetric scattering geometry using an argon ion laser ($\lambda_0 = 514.5 \text{ nm}$) and a low input laser power ($< 30 \text{ mW}$ focused to $\sim 30 \mu\text{m}$ diameter) in order to avoid back-transformation of the perovskite sample to enstatite or glass. The scattered light was analyzed by a six-pass Fabry-Perot interferometer, and sound velocities were determined from the measured frequency shift (for further details, see *Sandercock* [1982] and *Sinogeikin et al.* [1998]). To reduce geometric or other possible systematic errors in velocity measurements, the Brillouin system was calibrated using an MgO single-crystal standard. A total of 12 spectra were

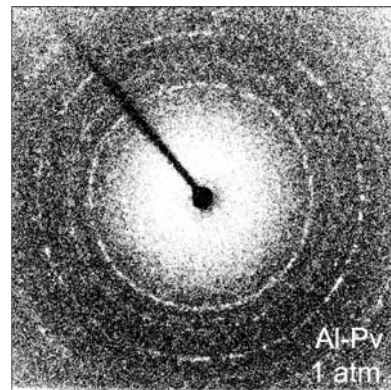


Figure 1. A monochromatic x-ray diffraction pattern of the polycrystalline Al-Pv sample used in this study. The sample was not rotated in the x-ray beam, and note that complete diffraction rings are nonetheless clear.

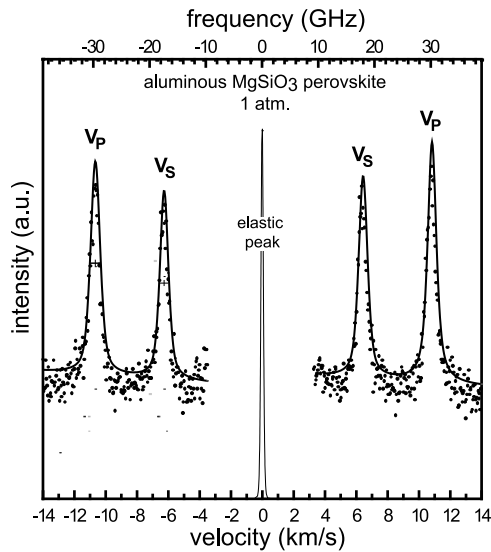


Figure 2. Brillouin spectrum of polycrystalline MgSiO_3 perovskite with 5.1 ± 0.2 wt.% Al_2O_3 . Solid circles are the observed data, and the compressional (V_P) and shear (V_S) acoustic modes of Al-Pv are labeled. The black curves are fits to the data (see text). The crosses represent the peak centers and the full width at half maximum (FWHM) for Al-Pv ($\text{FWHM}_{V_P} = 700$ m/s and $\text{FWHM}_{V_S} = 660$ m/s). The elastic peak has been scaled down and has a FWHM of 170 m/s. Conversion of the top scale in frequency (GHz) to the bottom scale in velocity (km/s) requires knowledge of the momentum transfer (i.e., the scattering angle) and can be written in the form: $V = 1/2 \Delta\nu_B \lambda_0 / \sin(\theta/2)$, where V is the phonon velocity, $\Delta\nu_B$ is the measured Brillouin frequency shift, λ_0 is the incident laser wavelength (514.5 nm), and θ is the angle between the incident and scattered light (90-degrees).

collected for Al-Pv from different areas of the sample and in different orientations. For each spectrum, the compressional (V_P) and shear (V_S) acoustic peaks (Stokes and anti-Stokes contributions) were fit with a mixed Gaussian plus Lorentzian profile. The standard deviations for V_P and V_S are both approximately 50 m/s, calculated from the variation in position of the peak centers for the 12 spectra. We emphasize that the scatter in peak positions is very small, with the 50 m/s root-mean-square variation close to the intrinsic instrumental variability. We also obtained a stacked spectrum by aligning the individual spectra on the zero-velocity-shift elastic peak position and summing them. The acoustic mode peak shapes in the stacked spectrum are symmetric and were fit with the same mixed Gaussian-Lorentzian profile as described above (Figure 2). This resulted in peak centers (reported in Table 1) that are within 50 m/s of the average of the individual spectra.

3. Results and Discussion

3.1. Sound Velocities of Al-Pv

[7] In order to assess the effect of aluminum on the elastic properties of Mg-Pv, the properties of Mg-Pv must be accurately known. Recent Brillouin scattering measurements on single-crystal and polycrystalline Mg-Pv are reported in *Sinogeikin et al.* [2004] and supercede those of previously reported single-crystal Brillouin results

[*Yeganeh-Haeri*, 1994] (for further details, see *Sinogeikin et al.* [2004]). We note that the aggregate sound velocities from the experiments using polycrystalline and single-crystal MgSiO_3 perovskite agree within their respective 1σ uncertainties (Table 1), and the bulk moduli obtained in these Brillouin experiments are in agreement with the recent static compression results of *Fiquet et al.* [2000]. Comparing our results on Al-Pv with the results of *Sinogeikin et al.* [2004] and the ultrasonic results of *Sinel'nikov et al.* [1998], we find that the compressional wave velocity of Al-Pv is indistinguishable from that of Mg-Pv, whereas the shear wave velocity is decreased by 2.4% (Table 1). The adiabatic bulk modulus (K_S) and shear modulus (μ) were calculated from the measured compressional and shear acoustic modes using the relations: $K_S = \rho(V_P^2 - 4/3V_S^2)$ and $\mu = \rho(V_S^2)$. The associated uncertainties were calculated using the standard error propagation method. Compared with the average of reported values for Mg-Pv, including the most recent P-V-T EOS for Mg-Pv proposed by *Fiquet et al.* [2000] (Table 1), we find that the bulk modulus of Al-Pv is indistinguishable from that of Mg-Pv, within experimental uncertainties. However, the shear moduli of these materials exhibit a significant difference. We find a 5.6% reduction in μ , showing that aluminum is remarkably effective in softening the shear properties of magnesium silicate perovskite. In fact, Al appears to have a greater effect than Fe on the shear modulus of Mg-Pv [*Kiefer et al.*, 2002].

3.2. Aluminum Incorporation

[8] Theoretical simulations show that the compressibility of Al-Pv and the mechanism by which aluminum is incorporated into the perovskite crystal structure are related [*Brodholt*, 2000; *Yamamoto et al.*, 2003; *Akber-Knutson and Bukowinski*, 2004]. The incorporation of Al^{3+} may occur by two competing mechanisms: where Al substitutes for Mg and Si in equal amounts, $\text{Mg}_{\text{Mg}} + \text{Si}_{\text{Si}} \rightarrow \text{Al}_{\text{Mg}}^{3+} + \text{Al}_{\text{Si}}^{3-}$ (mechanism A, e.g., *Andraut et al.* [1998]; *Andraut et al.* [2001]; *Stebbins et al.* [2001]; *Yamamoto et al.* [2003]; *Akber-Knutson and Bukowinski* [2004]) or where Al enters the Si site only and oxygen vacancies are required for charge balancing, $\text{Si}_{\text{Si}} + 1/2\text{O}_{\text{O}} \rightarrow \text{Al}_{\text{Si}}^{3-} + 1/2\text{V}_{\text{O}}^{+2}$ (mechanism B, e.g., *Brodholt* [2000]; *Navrotsky et al.* [2003]; *Stebbins et al.* [2003]). The incorporation of Al via cation vacancies may also occur but is less likely [*Lauterbach et al.*, 2000]. Static first-principles DFT as well as atomistic calculations indicate that although both mechanisms may operate, mechanism A is energetically favored at all mantle pressures and results in a 1–2% reduction of the bulk modulus for Al-Pv, as compared to Mg-Pv, whereas a 4–9% reduction was calculated for mechanism B [*Yamamoto et al.*, 2003; *Akber-Knutson and Bukowinski*, 2004]. As discussed earlier, we find no clear evidence for non-stoichiometry in Al-Pv. Taken at face value, our results for the bulk modulus support the charge-coupled mechanism A [*Yamamoto et al.*, 2003; *Akber-Knutson and Bukowinski*, 2004]. Thus, the presence of Al, not oxygen vacancies, has a major control on shear elastic softening of Mg-Pv. If vacancies are present in our sample, they do not affect the bulk modulus or the equation of state as much as previously thought [*Zhang and Weidner*, 1999; *Brodholt*, 2000].

[9] Our results are in contrast to previous claims of about a 10% reduction in K_{OT} caused by the presence of Al [*Zhang*

and Weidner, 1999; Kubo et al., 2000; Daniel et al., 2001]. Our observations also do not support an increase of the bulk modulus with increasing Al content [Andraut et al., 2001]. The difference in values obtained for the bulk modulus of Al-Pv could be attributed to different sample synthesis conditions, the presence of non-hydrostatic conditions in previous compression experiments [Daniel et al., submitted manuscript, 2004], differences in experimental P-T conditions which could affect defect concentrations and therefore the results of compression and Brillouin experiments [Andraut et al., 2001], or errors associated with the pressure standards (e.g., NaCl, Pt, and Au) used in previous diffraction experiments. We note that the Al-Pv sample used in our study was synthesized under similar P-T conditions, and with a similar aluminum content, as the sample used by Zhang and Weidner [1999], yet we obtain significantly different values for the bulk modulus (see Table 1). Therefore, sample synthesis cannot explain these differences, and it is possible that the difference could arise in part from the equation of state used for fitting the P-V-T data.

3.3. Aluminum Heterogeneity in the Lower Mantle

[10] Recent seismic tomography images have shown evidence for lateral velocity variations in Earth's lower mantle [e.g., Grand and van der Hilst, 1997; van der Hilst and Karason, 1999; Masters et al., 2000; Montelli et al., 2004] that may be thermal or chemical in origin. In order to evaluate the effect of lateral variations in aluminum content, we calculate the relative variations of shear and compressional velocities (expressed as $R = \partial \ln V_s / \partial \ln V_p$) of Al-Pv and Mg-Pv. Using the sound velocities reported in Table 1, we find that 5.1 ± 0.2 wt.% Al_2O_3 in Mg-Pv results in $R = 2.3$ at ambient conditions. This value is close to that which is observed at shallow lower mantle depths (~ 1000 to 1700 km), but not as high as the value observed near the base of the lower mantle ($R = 3.7$) [Masters et al., 2000]. For comparison, DFT calculations show that variations of up to 25 mol% iron in Mg-Pv yield $R = 1.46$ at zero pressure and $R = 1.6$ at 136 GPa [Kiefer et al., 2002]. Our results would indicate that variations in aluminum content of silicate perovskite is a possible candidate for causing lateral velocity heterogeneity in at least the uppermost portion of the lower mantle. If plumes rise from the lower mantle [Montelli et al., 2004] containing an enhanced basaltic component, then the large variations in R observed in the central Pacific and Africa (for example) could be due to lower mantle heterogeneity related to variations in the basaltic component of lower mantle material.

[11] **Acknowledgments.** We are grateful to Ian Steele for the microprobe analyses and to Wolfgang Sturhahn, Ian Steele, S. V. Sinogeikin, and I. Daniel for helpful discussions. This research was supported by NSF grant EAR 0003383 and 0135642. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the National Science Foundation.

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