Temperature derivatives of elastic moduli of $MgSiO_3$ perovskite

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Abstract. Elastic properties of polycrystalline $MgSiO_3$ perovskite were determined by means of resonance sphere technique at temperatures from 258 to 318 K under atmospheric pressure. The temperature derivatives of adiabatic bulk $(\partial K_S/\partial T)_P$ and shear moduli $(\partial G/\partial T)_P$ are –0.029(2) GPa/K and –0.024(1) GPa/K, respectively. Applying the present results in interpretation of seismological observations, we suggest that the chemical composition of the earth's lower mantle is pyrolitic at the potential temperature of 1600 K.

1. Introduction

It is widely recognized that Mg-rich silicate perovskite is the most abundant mineral in the earth's lower mantle [e.g., *Ito and Takahashi*, 1989; *Jeanloz and Knittle*, 1989; *Jackson*, 1998]. Therefore, its elastic properties are of prime importance to constrain the chemical composition and temperature profile in the lower mantle. The elastic moduli of silicate perovskite at ambient conditions have been determined with Brillouin scattering [*Yeganeh-Haeri*, 1994]. However, the pressure and temperature derivatives of the elastic moduli are poorly constrained. Most previous experiments using diamond anvil cell were suitable for obtaining a static compression curve yielding only isothermal bulk modulus (K_T), its temperature derivative, and thermal expansion (α) with relatively large uncertainty [e.g., *Mao et al.*, 1991; *Wang et al.*, 1994; *Funamori et* *al.*, 1996; *Fiquet et al.*, 2000].

Sinelnikov et al. [1998] reported the pressure and temperature derivatives of shear modulus using elastic wave velocity measurements at pressures as high as 8 GPa and temperatures up to 800 K. Their temperature derivatives, however, were derived from the shear moduli measured at various *P-T* conditions. Therefore, their results may be constrained by the uncertainties not only of temperature but also of pressure.

In this study, we conducted measurements to obtain temperature derivatives for both bulk and shear moduli of polycrystalline $MgSiO_3$ perovskite using resonant sphere technique [*Suzuki et al.*, 1992] at atmospheric pressure. Relatively high precision in determining temperatures and resonance frequencies in this technique enabled us to determine the temperature derivatives of the elastic moduli even for small temperature variations.

2. Experimental

A polycrystalline sample of $MgSiO_3$ perovskite was synthesized with the Kawai-type high-pressure apparatus at pressures and temperatures around 23 GPa and 1800 K for a run duration of 1 hour. The recovered sample was confirmed to be pure aggregate of perovskite by micro-focus X-ray diffraction spectra. It was carefully made into a spherical shape by means of the two-pipe method [*Fraser and LaCraw*, 1964] with a diameter of 1.303 (6) mm. The bulk density was determined to be 4.10(6) x 10^3 kg/m^3 which was close to 4.108 x 10^3 kg/m^3 taken from the literature [*Ito and Matsui*, 1979].

The resonance spectra were acquired from 3.8 to 8.0 MHz at 10 K intervals between 258 and 318 K. An example of the spectra is shown in Figure 1. Nine resonant modes ($_0T_2$, $_0S_2$, $_1S_1$, $_0T_3$, $_0S_3$, $_0S_0$, $_1S_2$, $_0S_4$, $_0T_4$) were recognized. However, the peaks of toroidal

(${}_0T_2$, ${}_0T_3$ and ${}_0T_4$) modes are highly sensitive to the distortion in the sample shape from the sphere, which results in the substantial peak splitting (Figure 1). Therefore, we omitted those peaks and also adjacent spheroidal modes (${}_0S_3$, ${}_0S_4$) which can not be distinctively recognized. The rest of 4 spheroidal modes (${}_0S_2$, ${}_1S_1$, ${}_0S_0$, ${}_1S_2$), being less sensitive to shape irregularity and thus showing slight peak splitting, were used in the analyses followed. It should be noted that even if we considered the average value of the substantially scattered but identified peaks (${}_0T_2$ mode) which was omitted from the consideration, the resulting variations are as small as ~1 % for absolute value and within ~6 % for the temperature derivatives. At ambient pressures, it is known that silicate perovskite becomes amorphous at around 400 K [*Wang et al.*, 1994]. The highest temperature of the present measurements was 318 K, low enough to avoid amorphization of the sample. Although the temperature variation was limited to be only 60 K, the temperature was controlled within 0.1 K and we observed the resonant frequencies clearly shifted with changing temperatures (Figure 2).

Results and Discussion

The elastic wave velocities (V_P and V_S) at 298 K were determined to be 10.7 and 6.51 km/s, respectively, using a least-square analysis based on the results obtained from four spheroidal modes. The results were consistent being within 3 % of the commonly accepted data calculated from Brillouin scattering (Table 1). The bulk and shear moduli were computed from those velocities for each temperature interval (Figure 3). The temperature derivatives of elastic moduli were calculated with a linear fitting (Table 1). The errors at each data point are mostly caused by the uncertainty of the diameter. Therefore, the errors of the temperature derivatives are not constrained by them, but instead, reflect the variations of resonant frequencies at each measurement. The temperature derivative for shear modulus thus obtained ($(\partial G/\partial T)_P$ = -0.024(1) GPa/K) was somewhat smaller in magnitude than the data by the ultrasonic measurements (-0.029(3) GPa/K) [*Sinelnikov et al.,* 1998]. On the other hand, the temperature derivative of K_S ($(\partial K_S/\partial T)_P$ = -0.029(2) GPa/K) was in good agreement with the temperature derivative of K_T determined by the static compression [*Funamori et al.,* 1996]. Although the shear modulus (G=174.1(5)) agreed well with other studies [*Yeganeh-Haeri,* 1994] within 2% (Table 1), the bulk modulus (K_S=237.7(8)) differed by 4 to 10% compared with the literature values as shown in Table 1 [*Yeganeh-Haeri*, 1994; *Andrault et al*., 2001]. The discrepancy is mainly caused by disagreement in V_P. Relatively low K_S in the present study suggests that a small amount of pores exists within the specimen. Even if the present specimen may be porous, however, it has been recognized that shear moduli in resonance experiments are less sensitive to the porosity compared with bulk moduli [*Suzuki et al*., 1992; *Katsura et al*., 2001].

Previous ultrasonic experiments at high pressures (~8 GPa) [*Sinelnikov et al*., 1998] were conducted in the temperature range of up to 800 K. The ultrasonic velocity measurements were performed in a solid pressure-medium and at various *P-T* conditions outside the thermodynamic stability field of $MgSiO_3$ perovskite. Therefore, they had difficulty in determining the temperature derivatives at constant pressure, which requires corrections of the pressure fluctuations associated with temperature change. In addition, in a solid pressure-medium, the effect of deviatoric stresses causes the uncertainty of pressure, which may affect the ultrasonic velocity measurement at high pressure.

We calculate the V_P and V_S for possible mantle models at high temperature and atmospheric pressure and compare them with the values inferred for the adiabatically decompressed lower mantle [*Jackson et al*., 1998]. The lower mantle is assumed to be a mixture of perovskite (*Pv*) and magnesiowüstite (*Mw*) with X_{Pv} ranging from 0.67 to 1.0 (X_{Mw}=0.33-0) in molar ratio. The total iron content, Fe/(Fe+Mg) was fixed at 0.12. The partition coefficient (k) of Fe and Mg between *Pv* and *Mw* phases represented by $k=(X_{Mg}/X_{Fe})^{Pv}/(X_{Mg}/X_{Fe})^{Mw}$. Because the experimentally determined values of k range widely, we adopted 0.25 [*Katsura and Ito,* 1996], and somewhat higher value 0.45 [*Kesson e al.,* 1998] is also employed in the following analysis. As the elastic moduli (M_0) of perovskite phase, we used K_S =260 (±5) and G=177 GPa, which are commonly accepted values. Temperature corrections were applied to the elastic properties by $M=M_0+ (\partial M/\partial T)_P \cdot (T_p-300)$ and to the density of both phases based on the present results, together with recent studies [e.g., *Trampert et al*., 2001; *Kung et al*., 2002]. The elastic wave velocities of the lower mantle are computed at a potential temperature (T_p) of 1600 K, which is plausible in order to account for the depths of major seismic discontinuities interpreted as phase transitions [*Ito and Katsura*, 1989; *Jackson,* 1998].

Our model calculations at 1600 (±100) K (Figure 4) demonstrate that the elastic wave velocities of pyrolitic composition (X_{Pv}=0.67) are preferable in order to account for those estimated from the seismological observations [*Dziewonski and Anderson,* 1981; *Kennett et al*., 1995], even if the effect of Fe-Mg partitioning is considered. On the other hand, pure perovskite model yields velocities too fast to reconcile with the observations. Unlike the estimates based on the previous results of the temperature derivatives [*Sinelnikov et al*., 1998], which favors the more perovskite-enriched composition than pyrolite (X_{Pv}=~0.75 in Fig. 4), the present results suggest uniform chemical composition throughout the mantle. In order to provide a tighter constraint for the chemical and thermal model of the lower mantle, the effect of Fe and Al on the elasticity of silicate perovskite should be considered [*Zhang and Weidner*, 1999; *Andrault et al.*, 2001; *Kiefer et al.*, 2002] in the future.

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Figure captions

Figure 1. An example of resonant spectra at 258 K. Data was obtained at 100 Hz intervals.

Figure 2. Temperature dependence of resonant frequencies in $MgSiO_3$ perovskite for ${}_0S_2$ and ${}_0S_0$ resonant modes. The temperature was controlled within 0.1 K during the measurements at 10 K intervals. Circles and diamonds represent heating and cooling processes, respectively.

Figure 3. Adiabatic bulk and shear moduli of $MgSiO_3$ perovskite as a function of temperature. Relatively large errors resulted from uncertainty of the diameter.

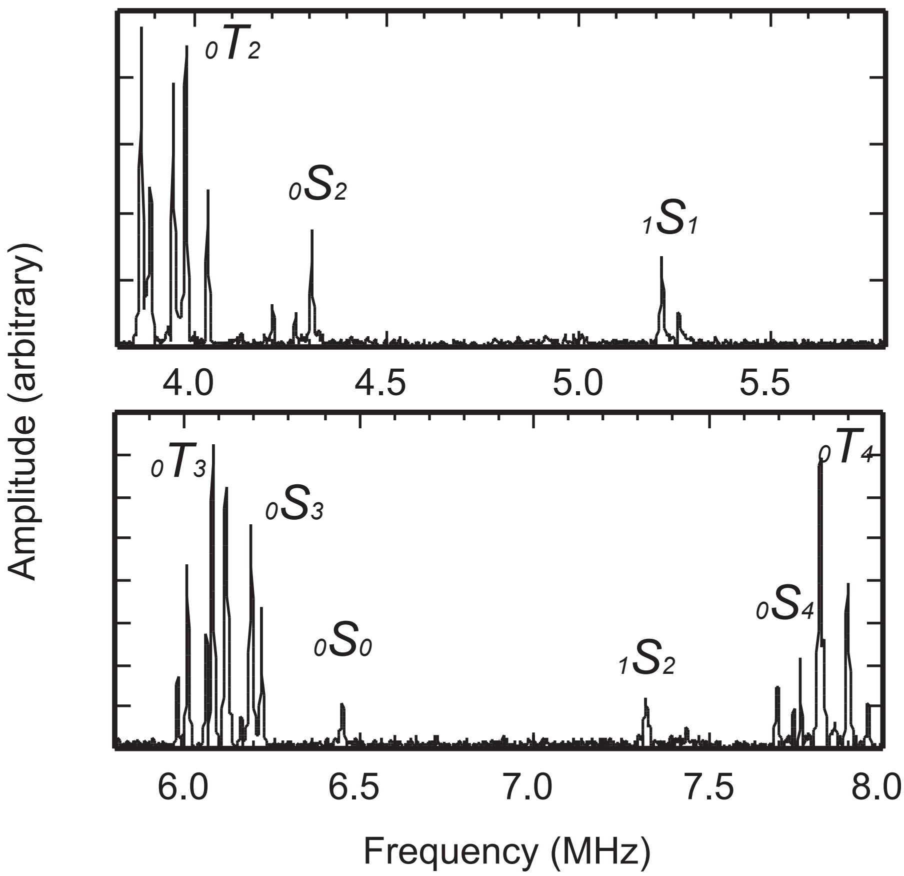
Figure 4. Estimated elastic wave velocities of (Mg, Fe)SiO_3 perovskite plus (Mg, Fe)O magnesiowüstite mixtures at around 1600 K at atmospheric pressure. For comparison, decompressed seismic wave velocities [Jackson, 1998] based on the seismological model [*Dziewonski and Anderson*, 1981; *Kennett et al.*, 1995] are shown by open square. Note that the temperature at atmospheric pressure designates potential temperature in the earth's interior. The velocities are calculated for various bulk compositions of X_{Pv} and at temperatures 1600±100 K. Solid black and gray diamonds represent the V_{P} and V_{S} for the model lower mantle (X_{Pv} = 0.67-1.0) with k of 0.25 and 0.45, respectively. Open diamonds exhibit the velocities calculated based on the previous study [*Sinelnikov et al.*, 1998]. Error bars correspond to the uncertainties of the potential temperature (±100 K) and the value of Ks (±5 GPa).

Table 1. Bulk and shear moduli and their temperature derivatives of magnesium silicate perovskite

Reference	K_S *(GPa)*	*K'*	*G (GPa)*	V_P *(km/s)*	V_S *(km/s)*	$(\partial K_T/\partial T)_P$	$(\partial G/\partial T)_P$
This study	237.7(8)		174.1(5)	10.70	6.51	-0.032(2)[#]	-0.024(1)
<a>	264(5)		177(3)	11.03	6.56		
<b>	249(3)	4.0fixed					
<c>	253(9)	3.9				-0.017(2)	
<d>	261(4)	4.0fixed				-0.05(1)	
<e>	261	4.0fixed				-0.023(11)	
<f>	261	4.0fixed				-0.028(3)	
<g>			175(6)				-0.029(3)

<a> Brillouin-scattering measurements for $MgSiO_3$ single crystal; Yeganeh-Haeri, 1994; <b>-<e> Static compression in DAC; <b> Andrault et al., 2001; <c> Fiquet et al., 2000; <d>* Fe-bearing $MgSiO_3$ perovskite (Mao et al., 1991); <e> Wang et al., 1994; <f> Static compression in Kawai-type apparatus; Funamori et al., 1996; <g> Ultrasonic interferometry for polycrystalline samples; Sinelnikov et al., 1998.

[#] K_S is converted into K_T using the relation of $K_T=(1+\alpha\gamma T)K_S$, where γ is assumed to be constant, 1.3.



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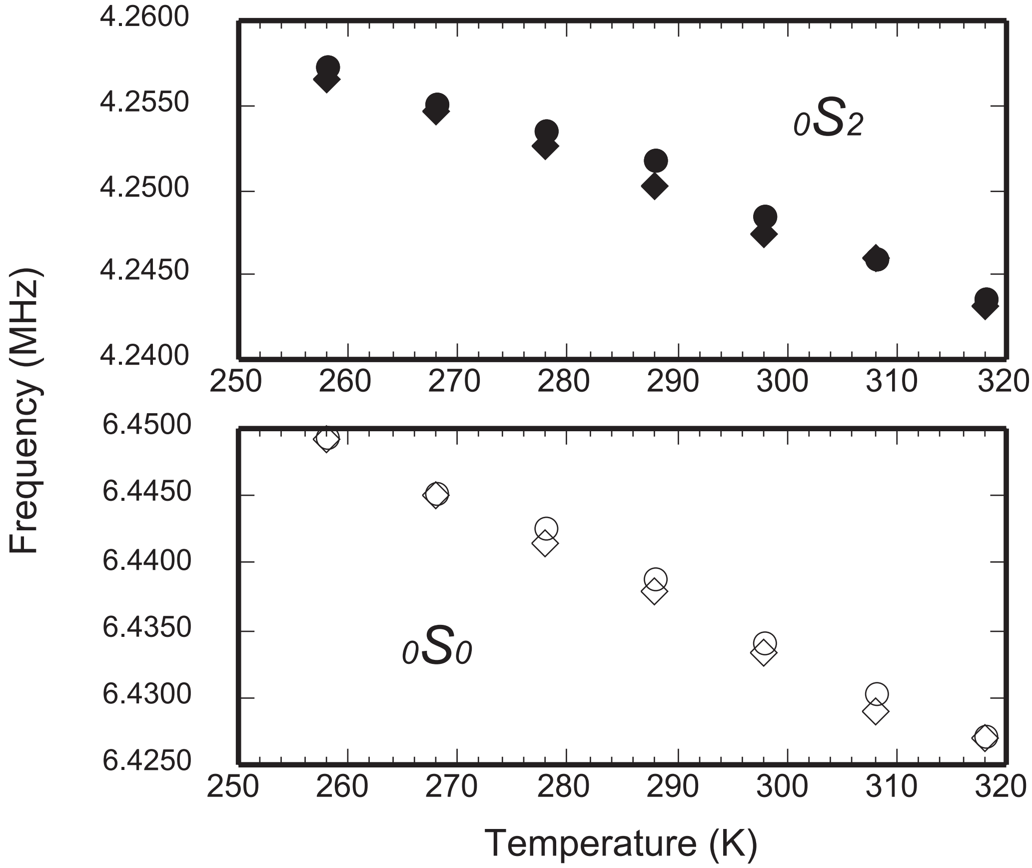

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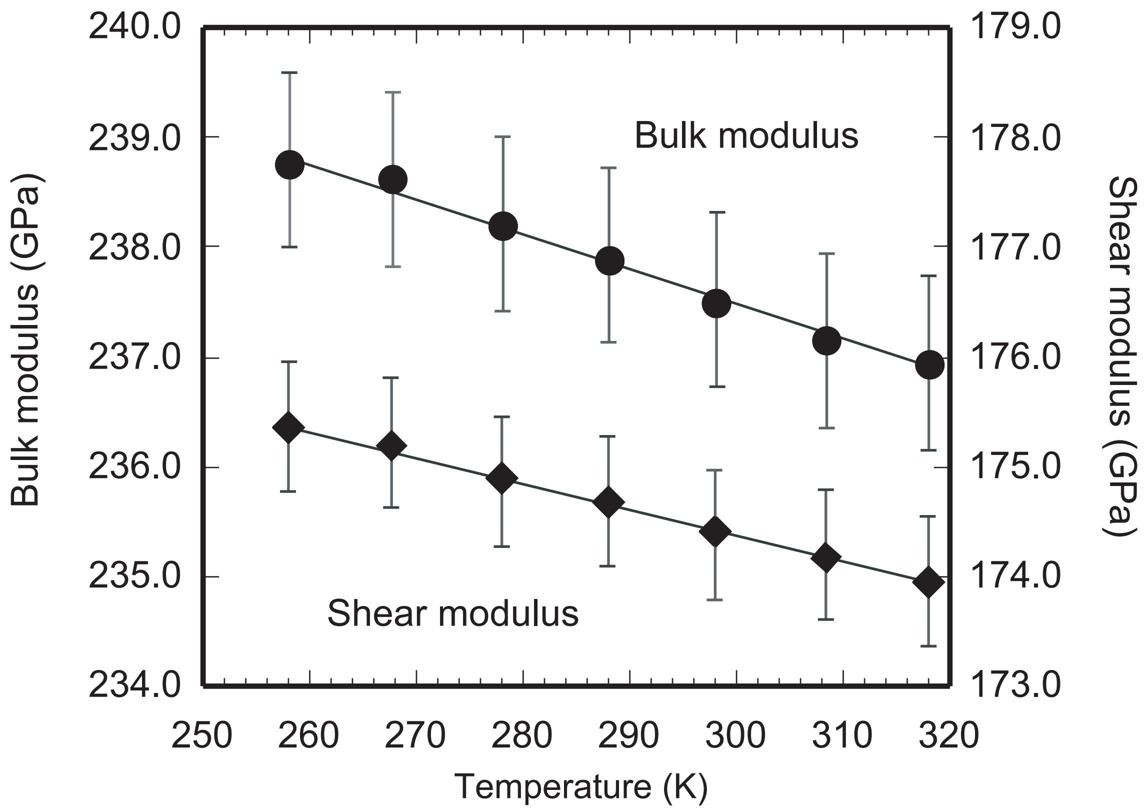

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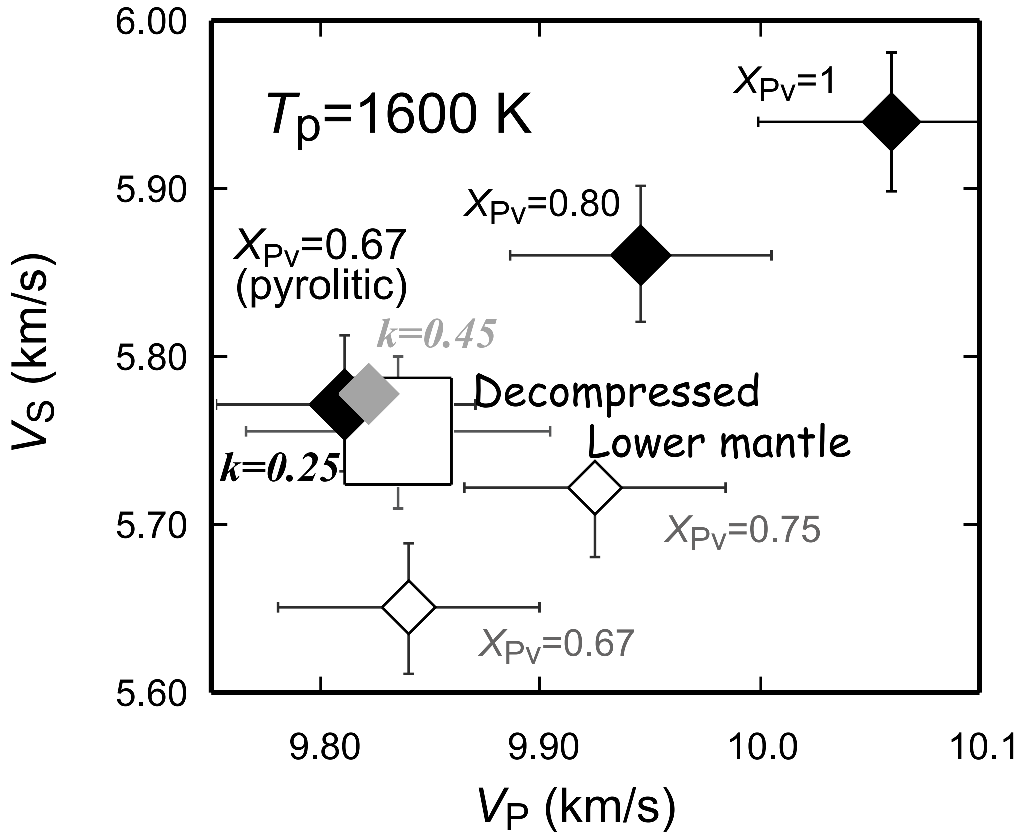

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