

Shifts in thermal expansivity with Fe content for solid solutions of $\text{MgSiO}_3\text{-FeSiO}_3$ with the perovskite structure

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ABSTRACT

This study presents evidence that for a solid solution of $\text{Mg}_x\text{Fe}_{x-1}\text{SiO}_3$ perovskite, the shift in volume thermal expansivity, α , is small as the index x is changed. According to data obtained theoretically by Hama and Suito (1998), α decreases by 0.3–0.4% as x changes from 1 to 0.9 in a temperature range of 1900 K. Furthermore, the relative shift in α for Fe substitution is –0.4% under lower mantle conditions. Hama and Suito used the Vinet equation of state to calculate thermoelastic properties. Using a thermodynamic approach applied to the properties of a Debye solid, the data show that the relative shift in α is less than the relative change in specific volume, and, as x changes from 1 to 0.9, V increases by 0.6% and α decreases by 0.4%.

INTRODUCTION

Figure 1 introduces the problem to be discussed here. The measured and calculated volume thermal expansivity, α , of MgSiO_3 perovskite (Fe-free perovskite, or simply Pv) lies in a band in the lower part of the graph, whereas the two high-temperature (T above 1000 K) measurements of α of Fe-rich perovskite, $(\text{Mg}_{0.9}\text{Fe}_{0.1})\text{SiO}_3$ (Fe-Pv), lie considerably higher on the graph. The dichotomy shown in Figure 1 is somewhat oversimplified because the experimental data in the lower band have different T ranges of measurement. One curve in the lower band represents an extrapolation of data measured by Wang et al. (1994). Yet another curve in the lower band represents an independent theoretical calculation (for more details, see Fig. 1 caption). Nevertheless, Figure 1 correctly demonstrates the disparity between the high $\alpha(T)$ values held by one group of authors and the low $\alpha(T)$ values held by other groups of authors. At high T , Fe-Pv has an α value 1.5 times that of Pv. The Fe-Pv set of α curves produces a density profile at mantle conditions matching that of PREM, suggesting a chondritic-like lower mantle composition (Stixrude et al. 1992; Hemley et al. 1992; Jeanloz and Knittle 1989). The Pv α curves do not generate lower mantle density curves matching those of PREM, suggesting a pyrolytic composition (Jackson 1983; Wang et al. 1994; Zhao and Anderson 1994; Chopelas 1996; Jackson and Rigden 1996; Hama and Suito 1998). Such a large difference between these two sets of solutions for α was unexpected five years ago because values of the bulk modulus (K_T) and its pressure derivative (K'_T) were essentially the same for Fe-Pv as for Pv. This led to predictions that α would also not depend on Fe content, and

therefore, future measured data on Pv α would converge with those on Fe-Pv α [in particular the upper curves in Fig. 1 (Jeanloz and Hemley 1994)]. As seen in Figure 1, high T values of α found by recent measurements of MgSiO_3 do not converge with high T values of α for Fe-Pv found by Mao et al. (1991) and Knittle et al. (1986), as Jeanloz and Hemley (1994) had predicted.

In an attempt to reconcile the data in Figure 1, it could be conjectured that the high T values of α for Fe-Pv arise from the Fe itself (e.g., a considerable shift occurs in α with Fe content). A recent theoretical paper shows such a conjecture to be unfounded. Hama and Suito (1998), solving for thermoelastic constants using the Vinet equation of state, demonstrated that Fe-Pv and Pv yield virtually the same value of α at the P and T conditions of the lower mantle (see their Fig. 11). The data behind Figure 11 of Hama and Suito (1998) are listed in Table 1 here.

Thermodynamically, this study shows that the shift in α going from Pv to Fe-Pv is small by exploiting the properties of a Debye model. MgSiO_3 perovskite has been proven to be a Debye-like solid by showing that using the Debye theory for Pv produces values of four thermoelastic parameters that agree with those measured or calculated by others (Anderson 1998). Further proof is shown in Table 2, which lists the values of K_T (isothermal bulk modulus), K_S (adiabatic bulk modulus), v_p (longitudinal sound velocity), v_s (shear velocity), and γ (Grüneisen parameter), all vs. T , from both the Anderson (1998) Debye calculation and the Hama and Suito (1998) calculation. This table shows general agreement between the results from the Debye model and those from the theory based on the Vinet equation of state. The calculated volumetric thermal expansion of Fe-Pv α data is in good

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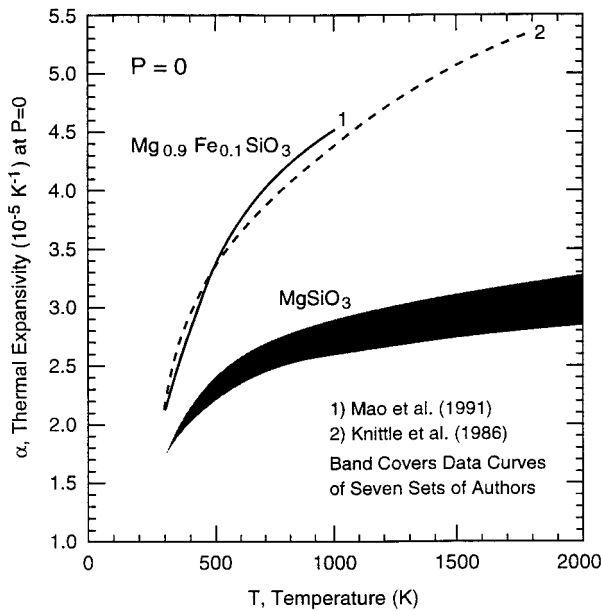


FIGURE 1. Thermal expansivity, α , vs. T curves. The banded section represents the measured and calculated curves for MgSiO_3 perovskite. The upper two curves represent measurements of $\text{Mg}_{0.9}\text{Fe}_{0.1}\text{SiO}_3$ perovskite: (1) as taken by Knittle et al. (1986) and reported by Saxena et al. (1993) in a table labeled “ MgSiO_3 Perovskite”, and (2) as measured by Mao et al. (1991) and graphed by Chopelas (1996). Note the large difference between α curves for the MgSiO_3 perovskite, and those for Fe-rich perovskite, especially pronounced at high T . The banded section includes $\alpha(T)$ curves from Wang et al. (1994), Funamori et al. (1996), Chopelas (1996), Gillet et al. (1996), Jackson and Rigden (1996), Anderson et al. (1996), and Hama and Suito (1998) (modified from Anderson 1998, Fig. 8). Of these curves in the banded zone, two (from Wang et al. 1994 and Funamori et al. 1996) have data over a wide T range, two (from Chopelas 1996 and Gillet et al. 1996) have data over a limited T range, and two (from Anderson et al. and Hama and Suito) are theoretical.

agreement with experimental values for 150–500 K at $P = 0$ (Hama and Suito 1998).

THE BULK MODULUS OF A DEBYE-LIKE SOLID

The agreement of K_0 for MgSiO_3 and $\text{Mg}_{0.9}\text{Fe}_{0.1}\text{SiO}_3$ is expected for a Debye-like solid. Shankland (1972) showed in that a Debye solid in which the volume of the crystallographic cell does not change significantly with

cation substitution, the variation of bulk sound velocity, v_b , with density, ρ , obeys the rule

$$\frac{v_b}{v_{b0}} = \left(\frac{\rho}{\rho_0} \right)^{-1/2}. \quad (1)$$

Using the definition $v_b = \sqrt{K_s/\rho}$, where K_s is the adiabatic bulk modulus, at $P = 0$, Equation 1 becomes

$$K_s = \text{constant} \quad (2)$$

as ρ in the solid solution changes.

Equation 2 has been verified for silicate solid solutions with limited Fe content, including orthopyroxenes, olivines, and the MgO-Fe join (see Part 5.7.4 of Anderson 1995, p. 134–136). From Shankland’s theory and experiments involving solid solution series, we conclude that the bulk modulus will not change as Fe content changes in magnesium silicate perovskite, a finding emphasized by Jeanloz and Hemley (1994).

The bulk modulus, $V(\partial^2 F/\partial V^2)_T$ is derived from the static term E_{ST} in the Helmholtz free energy (Landau and Lifshitz 1958) of a dielectric non-magnetic solid

$$F = E_{\text{ST}} + \frac{1}{2} \sum_{j=1}^{3pN} \hbar\omega_j + kT \sum_{j=1}^{3pN} \ln(1 - e^{-\hbar\omega_j/kT}) \quad (3)$$

\hbar and k are Planck’s constant and the Boltzmann constant, respectively; N is Avogadro’s number; and p is the number of atoms in the simplest vibrating unit (2 for MgO , 5 for MgSiO_3). The last term on the right is the vibrational energy, and the term preceding it is the zero point energy. Hofmeister (1991, Eq. 32) showed that the static term leads to K_T being a complex function of masses and interatomic distances multiplied by $\sum v_i^2/V$. This term is calculated at 300 K, not $T = 0$. The finite temperature term needed to account for this approach is small (Anderson 1995, Eq 2.43), often exactly zero. The zero point energy contribution is also small but not negligible (Hofmeister 1997). Calculation of K_T for MgSiO_3 end-member from vibrational spectroscopy (Lu et al. 1994) yielded a value within the experimental uncertainty of compressional measurements of Mao et al. (1991). The frequencies measured for Fe-Pv by Lu et al. (1994) are only slightly reduced, and mostly the lowest lying modes are affected, which contribute the least to K_T . Because of the above and the fact that volume increase opposes the frequency increase (Hofmeister 1991, eq. 32), it is clear that bulk modulus of silicate perovskite will be unaffected by

TABLE 1. Comparison of α for MgSiO_3 and α for $\text{Mg}_{0.9}\text{Fe}_{0.1}\text{SiO}_3$ vs. T at $P = 0$

T (K)	α ($10^{-5}/\text{K}$)		T (K)	α ($10^{-5}/\text{K}$)	
	MgSiO_3	$\text{Mg}_{0.9}\text{Fe}_{0.1}\text{SiO}_3$		MgSiO_3	$\text{Mg}_{0.9}\text{Fe}_{0.1}\text{SiO}_3$
400	1.936	1.930	1400	2.792	2.781
600	2.301	2.294	1600	2.877	2.865
800	2.484	2.476	1800	2.962	2.949
1000	2.606	2.597	2000	3.051	3.035
1200	2.704	2.693	2200	3.143	3.126

Note: Data from calculations found in Hama and Suito (1998).

TABLE 2. Comparison of calculated MgSiO₃ perovskite properties

T (K)	K_T (GPa)		K_S (GPa)		v_p (km/s)		v_s (km/s)		γ	
	H & S	A	H & S	A	H & S	A	H & S	A	H & S	A
400	254.1		256.6	264.0	10.60	10.97	6.10	6.53	1.281	1.41
600	249.1	255.4	253.5	262.0	10.54	10.86	6.05	6.45	1.286	1.39
800	243.9	250.2	250.1	258.0	10.47	10.77	6.00	6.38	1.291	1.40
1000	238.5	244.9	246.5	254.2	10.41	10.66	5.95	6.95	1.297	1.40
1200	233.0	239.5	242.8	250.2	10.34	10.60	5.90	6.23	1.303	1.38
1400	227.4	234.0	239.1	246.9	10.27	10.45	5.85	6.15	1.309	1.38
1600	221.8	228.5	235.2	243.2	10.19	10.34	5.79	6.08	1.316	1.40
1800	216.1	223.0	231.3	239.5	10.12	10.23	5.73	5.73	1.323	1.41

Notes: H & S = Hama and Suito (1998); A = Anderson (1998).

small amounts of Fe. From the formulas in Hofmeister (1991), derivatives dK_T/dP and dK_T/dT would similarly be independent of Fe at the 10 to 20% level.

Many physical properties apart from K_{T_0} , such as α , are highly dependent on vibrational energy. For α , the essential physics revolves around the question of whether the substitution of Fe for Mg in the perovskite structure substantially changes the vibrational energy. We see from Equation 3 that the behavior of K_{T_0} provides no guide to predicting the behavior of α . Note that if K_0 is unchanged by Fe substitution, then K'_0 is also unchanged by Fe substitution.

VIBRATIONAL ENERGY OF THE DENSITY OF STATES (VDOS)

At sufficiently high temperatures, $kT \gg \hbar\omega$. The $\ln(1 - e^{-\hbar\omega/kT})$ term in Equation 3 is simplified so that the resulting high- T Helmholtz energy (Anderson 1995) becomes

$$F^{hr} = E_{ST} + kT \sum_{j=1}^{3pN} (\ln \hbar\omega_j - \ln kT) = E_{ST} + F_{TH}^{hr} \quad (4)$$

where F_{TH}^{hr} is the high T thermal energy. The thermal energy has absorbed the zero point energy, and consisting mainly of the vibrational energy, is the source of α , γ , $\partial K_T/\partial T$, δ_T , S , C_p , and all properties involving T derivatives of F^{hr} at constant V .

The total number of modes, $3pN$, will not change as pressure and temperature change in a solid solution in which cations are exchanged. But the properties of a lattice change when ω_j changes due to pressure or temperature. Assuming that the Debye density of states can be substituted for the VDOS, the Debye density of states will be used to find α of MgSiO₃ perovskite.

Changes in energy density of Fe-free MgSiO₃ perovskite (or Pv) may arise from changes in average interatomic distance due to cation substitution. Hama and Suito (1998) found the specific volume of Pv to be $V_0 = 160.14 \text{ \AA}^3$, while the ambient specific volume of Fe-rich Mg_{0.9}Fe_{0.1}SiO₃ (Fe-Pv variables hereafter designated with ') is $V'_0 = 161.16 \text{ \AA}^3$, an increase of 0.6%.

The maximum wave number in a Debye solid is $\mathbf{k}_m = \pi/r$, where r is the interatomic distance (Kittel 1956). Because r is proportional to $V^{1/3}$, we have $\mathbf{k}_m/\mathbf{k}'_m = (V_0/V'_0)^{1/3}$,

where the prime indicates that the silicate perovskite is enriched in Fe by the amount $\text{Fe}/(\text{Fe} + \text{Mg}) = 0.1$. Thus, $\mathbf{k}_m = 0.998 \mathbf{k}'_m$ resulting from a change in x from 1 to 0.9. In the Debye model, the frequency, ω , is proportional to k by $\omega = \bar{v}k$, so that $\mathbf{k}_m = \omega_m/\bar{v}$, where \bar{v} is the appropriate average sound velocity. In the treatment by Anderson (1998) of the Debye solid application to MgSiO₃ perovskite, \bar{v} is found from

$$\frac{3}{\bar{v}^3} = \left(\frac{2}{v_s^3} + \frac{1}{v_p^3} \right) \quad (5)$$

where v_s and v_p are the shear and longitudinal sound velocities. Using $\omega = \bar{v}k$,

$$\frac{\omega'_m}{\omega_m} = \frac{\bar{v}' k'_m}{\bar{v} k_m} = 0.998 \left(\frac{\bar{v}'}{\bar{v}} \right). \quad (6)$$

Note that \bar{v} serves as a function of ω . The equation for the density of states of a Debye solid involves ω^2 in the numerator and \bar{v}^3 in the denominator (Kittel 1956). Requiring the total number of modes of Fe-Pv to be equal to those of Pv leads to

$$\frac{\bar{v}'}{\bar{v}} = \left(\frac{\omega'_m}{\omega_m} \right)^{2/3}. \quad (7)$$

Placing Equation 7 in Equation 6, $(\omega'_m/\omega_m)^{5/2} = 0.998$ or $\omega'_m = 0.999\omega_m$. Because the Debye temperature is proportional to ω_m , we also have $\theta' = 0.999\theta$. Thus, the maximum frequency of the Debye density of states decreases by only 0.1% as a result of a change in x from 1 to 0.9. The value of ω_m of the VDOS has a similar decrease.

FINDING α FROM THE SPECIFIC HEAT

The definition of the Grüneisen parameter $\gamma = \alpha K_T V / C_v$ is used to find:

$$\frac{\alpha'}{\alpha} = \left(\frac{K_T}{K'_T} \right) \left(\frac{C'_v}{C_v} \right) \left(\frac{\gamma'}{\gamma} \right) \left(\frac{V}{V'} \right). \quad (8)$$

It has been shown that K_T and $(\partial K_T/\partial T)_{P=0}$ do not change with increasing Fe content for a Debye-like solid. Neither does γ change with increasing Fe content because at $P = 0$, γ is governed by $(\partial K_T/\partial P)_T = K'_0$ (Stacey 1995).

Thus:

$$\frac{\alpha'}{\alpha} = \left(\frac{C'_v}{C_v} \right) \left(\frac{V}{V'} \right). \quad (9)$$

Equation 9 shows that as V increases to V' , α' tends to diminish. Because an increase in V results in a decrease in θ , C_v decreases as V increases.

Using the Debye tables for C_v at $T \sim 1000$ K (Anderson 1995), decreasing θ by 0.1% increases C_v by 0.17% so that $C'_v/C_v = 1.0017$. Then α'/α is 0.996 considering that $V/V' = 1/1.006$. Thus α diminishes by 0.4% as x changes from 1 to 0.9, supporting the Hama and Suito (1998) results shown in Table 1. The small decrease in α results directly from the small increase in specific volume.

Therefore, the large difference between the sets of Fe-Pv α values and Pv α values in Figure 1 cannot be attributed to the changing Fe content. It has been shown that one of the two sets of data in Figure 1 is deviant. We now use the criterion of internal consistency to ascertain which data set is correct and which is deviant. The values of the thermoelastic parameters δ_T and γ for the upper set are high ($\delta_T = 7$ as found by Stixrude et al. 1992, and require $\gamma = 1.96$, as reported by Hemley et al. 1992). Anderson et al. (1996) showed that the high values of δ_T and γ found from the Mao et al. (1991), data by Stixrude et al. (1992), and Hemley et al. (1992) are thermodynamically inconsistent with the values of K_0 and K'_0 found by Mao et al. (1991). Therefore, the upper set of $\alpha(T)$ values in Figure 1 is the deviant set. The correct set gives $\delta_T = 5.0$ at 300 K and $\gamma = 1.5$ at 300 K (Anderson 1998), which are comparable to those of MgO and olivine.

CONCLUSIONS

Various authors have attempted to account for the unreasonably high (in their opinion) $\alpha(T)$ values shown in the top of Figure 1. Hama and Suito (1998) noted that Fe-Pv experimental data deviate from their EoS curves above about 700 K at each isobar (see their Fig. 9a). That is where the value of α begins to grow large in response to the sudden increase in the curvature of the V - T line shown by the data. Hama and Suito (1998) attributed this shift in V to metastability of Fe-rich MgSiO_3 . Wang et al. (1994) stated that their own "one bar volume data are reversible only up to 430 K. All of the data above 430 K are highly questionable. . ." Fei et al. (1996) showed that 10% Fe in perovskite is not stable below 1500 °C and will form magnesiowüstite and stishovite, and further, that stishovite assumes a lower Fe content.

Duba et al. (1997) demonstrated that synthesizing Fe-bearing perovskites with a large temperature gradient in a laser-heated diamond anvil cell results in creation of multiphase assemblages because of the large thermal gradient along with the constant f_{O_2} condition in such cells. Thus, speculation arises that the Mao et al. (1991) and Knittle et al. (1986) experiments were not done on Fe-rich perovskite, as assumed, but on some other material, perhaps a sampling of the multiphase assemblages proposed by Duba et al. (1997).

As Sherlock Holmes told a doubtful Watson, "when you have eliminated the impossible, whatever remains,

however improbable, must be the truth. . ." (Sir Arthur Conan Doyle, *The Sign of Four*, 1895).

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