Elasticity of Single-Crystal Olivine at High Pressures and Temperatures

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23 **Abstract**

24 Elasticity of single-crystal San Carlos olivine has been derived from sound velocity
25 and density measurements at simultaneous high pressure-temperature conditions up to
26 20 GPa and 900 K using in situ Brillouin spectroscopy and single-crystal X-ray
27 diffraction in externally-heated diamond anvil cells. These experimental results are
28 used to evaluate the combined effect of pressure and temperature on full elastic
29 constants of single-crystal olivine to better understand its velocity profiles and
30 anisotropies in the deep mantle. Analysis of the results shows that the shear moduli
31 display strong concave behaviors as a function of pressure at a given high temperature,
32 while the longitudinal modulus, C_{11} , and the off-diagonal moduli, C_{12} and C_{13} , exhibit
33 greater temperature dependence at higher pressures than at relatively lower pressures.
34 Using a finite-strain theory and thermal equation of state modeling for a pyrolitic
35 mantle composition along an expected mantle geotherm, our results show that the
36 magnitude of the V_P and V_S jumps at 410-km depth are 6% and 6.4%, respectively,
37 which are greater than that in seismic observations, suggesting a mantle olivine
38 content of 40-50 vol%, which is less than what is expected for the pyrolite model. Our
39 modeled velocity profiles for a metastable olivine wedge in the subduction slabs along
40 a representative cold slab geotherm are 6% and 10% lower than those of wadsleyite
41 and ringwoodite, respectively, at corresponding depths of the normal mantle. Our
42 modeled results also show that metastable olivine in the cold slabs could have strong
43 V_P and V_S anisotropies. The maximum V_P anisotropy is estimated to be 19-22% at
44 transition zone depth, whereas the maximum V_S splitting is 13-23% and increases

with depth. As a result, the presence of a metastable olivine wedge at the transition zone depth would exhibit a seismic signature of low velocity and strong seismic anisotropy which are consistent with recent seismic observations for various locations of the slabs and can be used as mineral physics constraints for future seismic detections of the metastable olivine wedges in the deep mantle.

Keywords: Elasticity of single crystal, San Carlos olivine, High pressure and temperature, Metastable olivine wedge, Mantle, Seismic anisotropy

1. Introduction

Seismological studies have provided some of the most direct information for understanding the physical properties of the Earth's deep interior (e.g. Dziewonski and Anderson, 1981; Kennett et al., 1995; Romanowicz, 1991; van der Hilst et al., 2007). Interpretation of seismic images and profiles requires detailed knowledge of the elasticity of major mantle minerals at relevant pressure-temperature (P-T) conditions (Bass et al., 2008; Cammarano et al., 2005a; Duffy and Anderson, 1989; Ita and Stixrude, 1992; Marquardt et al., 2009; Murakami et al., 2012). As one of the most abundant minerals in the Earth's upper mantle, olivine has the volume percentage of 60% in a pyrolitic mantle composition (Ringwood, 1975). Studying the elasticity of olivine at P-T conditions relevant to the Earth's mantle is of fundamental importance in constraining the composition and understanding the structure of the region.

67 Experimental studies on the elasticity of olivine can be dated back to 1930s when
68 Adams (1931) studied the compressibility of fayalite, an Fe-endmember olivine
69 (Fe_2SiO_4), at ambient temperature. With the development of high pressure techniques,
70 the elasticity of olivine was extensively studied at high pressures and 300 K or high
71 temperatures and 1 bar in the past few decades (Abramson et al., 1997; Duffy et al.,
72 1995; Webb, 1989; Zha et al., 1996, 1998). Although Liu et al. (2005) reported the
73 aggregate bulk and shear moduli of San Carlos olivine at simultaneous high P-T
74 conditions up to 8 GPa and 1073 K using ultrasonic interferometry technique, *in-situ*
75 high P-T measurements on the elasticity of single-crystal olivine remain unavailable.
76 Much of our understanding of upper-mantle seismic structures and mineralogy still
77 heavily relies on the extrapolated elasticity of the candidate upper-mantle minerals,
78 including olivine, enstatite, and garnet (e.g. Abramson et al., 1997; Chai et al., 1997;
79 Duffy and Anderson, 1989; Duffy et al., 1995; Jackson et al., 2007; Jiang et al., 2004a;
80 Li and Liebermann, 2007; Webb, 1989; Zha et al., 1996, 1998). It is widely accepted
81 that the olivine content in the upper mantle can be constrained by comparing the
82 velocity contrast across the olivine-wadsleyite phase transition to seismic observations
83 for the 410-km discontinuity (Cammarano et al., 2005b; Duffy et al., 1995; Liu et al.,
84 2005). In this case, the olivine content in the upper mantle is estimated to be 30-50
85 vol%, a value much lower than 60 vol% in the pyrolite model. Such a low olivine
86 content model challenges the traditional pyrolite compositional model and calls for an
87 answer for reliably understanding the upper mantle mineralogy (Cammarano et al.,
88 2005b; Duffy et al., 1995; Li and Liebermann, 2007; Liu et al., 2005). The difference

in the olivine content between these models could be caused by our limited knowledge of the combined effect of P-T on the elasticity of olivine and wadsleyite at relevant mantle conditions. Therefore, high P-T measurements on the elasticity of olivine and other candidate mantle minerals are needed to provide a better constraint on the upper mantle mineralogy.

A considerable number of seismic studies have shown that Earth's upper mantle is seismically anisotropic in various locations (e.g. Assumpcao et al., 2006; Ekstrom and Dziewonski, 1998; Long and Becker, 2010; Long and van der Hilst, 2005; Marone and Romanowicz, 2007). In the upper 200-250 km depth, shear waves with horizontal polarization travel faster than waves with vertical polarization. This difference between two shear waves $[(V_{SH}-V_{SV})/V_{Voigt} \text{ \%}]$ can reach up to 4% (Ekstrom and Dziewonski, 1998; Long and Becker, 2010; Marone and Romanowicz, 2007; Nettles and Dziewonski, 2008). Since olivine as a major mantle phase has been shown to be strongly anisotropic in both compressional (V_P) and shear wave (V_S) velocities, the observed anisotropy has been suggested to be caused by the preferred alignment of olivine fabrics (Jung et al., 2009; Karato et al., 2008; Mainprice et al., 2005). Knowledge of the elasticity of single-crystal olivine is thus the key to understanding the anisotropic seismic structures of the upper mantle (Mainprice, 2007; Mainprice et al., 2000; Nunez-Valdez et al., 2010; Zha et al., 1996). Furthermore, seismic anisotropies have also been observed in the subduction slabs in the transition zone and used as potential evidence for the presence of the metastable olivine wedge in the

111 region (Chen and Brudzinski, 2003; Liu et al., 2008; Sandvol and Ni, 1997; Tono et
 112 al., 2009). We note that the phase transition of olivine to ringwoodite has been
 113 proposed to be a potential mechanism for occurrence of the deep earthquakes (e.g.
 114 Green et al., 2010; Green and Houston, 1995; Kirby, 1995). Knowing the elasticity of
 115 single-crystal olivine at P-T conditions relevant to the cold slab geotherm is thus
 116 essential for deciphering the seismic properties of the metastable olivine wedge. To
 117 date, there is no data available for the elasticity of single-crystal olivine at
 118 simultaneous high P-T (Abramson et al., 1997; Speziale et al., 2004; Webb, 1989; Zha
 119 et al., 1996, 1998), significantly hindering our progress in understanding the observed
 120 seismic anisotropy in the upper mantle and subduction slabs in the transition zone.

121
 122 In this study, we have investigated the single-crystal elasticity of San Carlos olivine at
 123 simultaneous high P-T conditions using Brillouin spectroscopy combined with
 124 single-crystal X-ray diffraction (XRD) and externally-heated DACs (EHDAC) up to
 125 20 GPa and 900 K. Using our experimental results, we have constructed a new
 126 velocity model in a pyrolite mantle composition to explore the nature of the 410-km
 127 discontinuity. We have also modeled the velocity and anisotropy profiles of the
 128 metastable olivine wedge in the subduction slabs to better understand its potential
 129 seismic structures in the transition zone.

130

131 **2. Experimental details**

132 Natural single-crystal San Carlos olivine $[(\text{Mg}_{0.9}\text{Fe}_{0.1})_2\text{SiO}_4]$ from Arizona, USA, was

133 examined for the chemical composition using the electron microprobe at The
134 University of Texas at Austin. The single crystal was cut into two pieces orthogonal to
135 each other with random crystallographic orientations of (0.96,0.12,0.24) (platelet 1)
136 and (0.49,-0.87,-0.03) (platelet 2), instead of with the principle orientations. For
137 low-symmetry minerals such as the orthorhombic olivine with nine independent
138 elastic constants, a large number of velocity measurements from different
139 crystallographic planes are typically needed to fully determine the elastic moduli of
140 single crystals at a given P-T condition, though obtaining such measurements at
141 extreme P-T is difficult in terms of the sample pieces and collection time needed.
142 Compared to principle planes, crystal pieces in random orientations normally exhibit
143 two polarized shear waves and one compressional wave velocity, which can be
144 simultaneously used to greatly reduce the need for conducting extensive velocity
145 measurements in various orientations. Such velocity measurements from two random
146 orientations also provide excellent constraints on the values and uncertainties of the
147 derived single-crystal elastic moduli (Mao et al., 2007; Speziale et al., 2004). Both of
148 the randomly cut sample pieces were double-side polished to platelets of 30-35 μm in
149 thickness and broken into several small platelets with a size of $\sim 80 \times 100 \mu\text{m}$ for the
150 high P-T Brillouin and XRD measurements. Single-crystal XRD patterns were used to
151 determine the orientations, crystal structure, and density of the sample at
152 GeoSoilEnviroCARS (GSECARS) of the Advanced Photon Source (APS), Argonne
153 National Laboratory (ANL). The starting sample had an orthorhombic structure with a
154 density of $3.343(3) \text{ g/cm}^3$ at ambient conditions. Re-W alloy, which is more stable at

155 high P-T conditions than typical Re gasket, was used as the gasket material for high
156 P-T experiments. The gasket was pre-indented to a thickness of $\sim 65\text{ }\mu\text{m}$ by a pair of
157 diamonds of $500\text{ }\mu\text{m}$ culet size in an EHDAC, and a hole of $300\text{ }\mu\text{m}$ was subsequently
158 drilled and used as the sample chamber. One selected platelet from each representative
159 orientation of the olivine crystal was loaded into an EHDAC together with Pt powder
160 which served as the pressure calibrant at high P-T (Fig. 1) (Fei et al., 2007). A ruby
161 sphere of approximately $5\text{ }\mu\text{m}$ was also loaded into the cell and used as the pressure
162 indicator for loading a Ne pressure medium as well as for high-pressure and
163 room-temperature experiments. The temperature of the sample was determined by an
164 R-type thermocouple attached to one of the diamond anvils approximately $500\text{ }\mu\text{m}$
165 away from the diamond culet. The EHDAC was equipped with an alumina ceramic
166 heater that was coiled by two Pt wires of $200\text{ }\mu\text{m}$ in diameter and 30 cm in length
167 (Kantor et al., 2012).

168

169 High P-T Brillouin measurements were conducted at up to 14 GPa at three given
170 temperatures of 500 K , 750 K , and 900 K at GSECARS of the APS, ANL, while
171 complimentary high-pressure Brillouin measurements at room temperature were also
172 performed up to 20 GPa at the Mineral Physics Laboratory of The University of Texas
173 at Austin. To avoid potential oxidation of the diamond anvils and Pt wires at high
174 temperatures, Ar with 2% hydrogen was continuously blown into the EHDAC while
175 the cell was heated. A solid state laser with a wavelength of 532 nm and a power of
176 0.4 mW was used for the Brillouin measurements in which Brillouin spectra were

collected at the forward scattering geometry with an external scattering angle of 50° using a six-pass Sandercock tandem Fabry-Perot interferometer. The acoustic velocities of the Brillouin spectra were derived from the measured Brillouin frequency shift as follows:

$$v = \frac{\Delta v_B \lambda_0}{2 \sin(\theta/2)} \quad (1)$$

where v is the measured acoustic velocity, Δv_B is the Brillouin frequency shift, λ_0 is the laser wavelength of 532 nm, and θ is the external scattering angle. For each platelet, we have collected the spectra at a 10° step vertically rotating over a range of 180° in the azimuthal direction (Fig. 1). For platelet 2, two quasi-transverse modes were observed for most directions, whereas only one quasi-transverse mode was detected in platelet 1 (Figs. 1 and 2). For the high-temperature measurements, the pressure was determined from the X-ray diffraction patterns of the Pt powder before and after each Brillouin measurement (Fei et al., 2007). We have also collected X-ray diffraction patterns of olivine at each given high P-T condition to determine the density of the sample. For the 300 K measurements at The University of Texas at Austin, the ruby fluorescence line was used to determine the pressure (Mao et al., 1986).

3. Results

At a given P-T condition, nine single-crystal elastic constants of olivine were obtained by simultaneously fitting the velocity data of two sample platelets with varying azimuthal angles using the Christoffel's equation (Figs. 2 and 3, Table S1) (Every,

199 1980):

$$200 \quad | C_{ijkl} n_i n_j - \rho v^2 \delta_{ik} | = 0 \quad (2)$$

201 where v is the measured velocity, ρ is the density determined by single-crystal XRD,
202 and δ_{ik} is the Kronecker delta function. The single-crystal elastic constants, C_{ijkl} , are
203 written in full suffix notation in the equation, although we have used reduced notation,
204 C_{ij} , for our reported values (Table S1). n_i represents the direction cosine of the phonon
205 propagation direction, which can be described by three Eulerian angles (θ, χ, ϕ) and
206 determined using single-crystal XRD measurements.

207

208 At 300 K, all of the longitudinal and off-diagonal moduli follow a nearly linear
209 increase with pressure, whereas the shear moduli, C_{44} , C_{55} , and C_{66} , exhibit a
210 downward trend towards higher pressures (Fig. 3). The aggregate bulk (K_S) and shear
211 moduli (G) in the Voigt-Reuss-Hill average are then calculated using these
212 single-crystal elastic moduli (Figs 4 and 5). At ambient conditions, our Brillouin
213 measurements yield the adiabatic bulk modulus, $K_{S0}=129.8(9)$ GPa, and shear
214 modulus, $G_0=77.8(5)$ GPa, which are in excellent agreement with literature results
215 (Abramson et al., 1997). Although K_S increases almost linearly with pressure, G at
216 300 K exhibits a downward trend with pressure due to the softening of the shear
217 moduli, C_{44} , C_{55} , and C_{66} (Fig. 4 and 5). Elevating temperature at a given pressure
218 decreases all of the elastic moduli. In general, most of the elastic moduli exhibit
219 similar pressure dependence at the investigated temperature range (Fig. 3 and Table
220 S2). However, the off-diagonal moduli, C_{12} and C_{13} , show temperature-induced

reduction more significantly at higher pressures (Fig. 3). The longitudinal moduli, C_{11} , also displays similar temperature-dependent trend at higher pressure.

The P-T derivatives of the elastic moduli were obtained by fitting the experimentally-derived elastic constants using the third-order or fourth-order finite-strain equations as follows (Fig. 3 and Table S2) (Birch, 1978):

$$C_{ij0}(T) = C_{ij0}(300K) + (T - 300)(\partial C_{ij} / \partial T)_P \quad (3)$$

$$C_{ij} = (1 + 2f)^{7/2} [C_{ij0}(T) + a_1 f + a_2 f^2] + a_3 P \quad (4)$$

$$a_1 = 3K_{T0}(C'_{ij} - 3) - 7C_{ij0}(T) \quad (5)$$

$$a_2 = 9K_{T0}^2(T)C''_{ij} + K_{T0}(T)(9K'_{T0} - 48)(C'_{ij} - 3) + 63C_{ij0}(T) \quad (6)$$

$$f = (1/2)[(V_0/V)^{2/3} - 1] \quad (7)$$

where $C_{ij0}(300\text{ K})$ is the derived zero-pressure constant from ambient measurements and is thus fixed for the modeling, $C_{ij0}(T)$ is the elastic constant at high temperature and 1 bar, f is the finite strain, V is the volume at high P-T determined by single-crystal XRD, V_0 is the volume at 1 bar and high temperature, $C'_{ij} = (\partial C_{ij} / \partial P)_T$ is the pressure derivative of the elastic constants, $C''_{ij} = (\partial^2 C_{ij} / \partial^2 P)_T$ is the second pressure derivative of the elastic constants, $K_{T0}(T)$ is the isothermal bulk modulus at 1 bar and high temperature, $K'_{T0} = (\partial K_{T0} / \partial P)_T$ is the pressure derivative of the bulk modulus. The parameter a_3 is 3 for the longitudinal moduli, C_{11} , C_{22} , and C_{33} , and is 1 for the rest of the elastic moduli. $K_{T0}(T)$ and K'_{T0} are calculated from the adiabatic bulk modulus K_{S0} and K'_{S0} as follows:

$$K_{T0}(300) = K_{S0} / (1 + \alpha \gamma T) \quad (8)$$

$$K_{T0}(T) = K_{T0}(300) + (\partial K_T / \partial T)_P (T - 300) \quad (9)$$

$$K'_{T0} = (1 + \alpha\gamma T)^{-1} [K'_{S0} - \gamma T / K_{T0}(T) (\partial K_T / \partial T)_P] \quad (10)$$

$$(\partial K_T / \partial T)_P \cong (\partial K_S / \partial T)_P / (1 + \alpha\gamma T) - K_S(T) / (1 + \alpha\gamma T)^2 [\alpha\gamma + (\partial \alpha / \partial T) \gamma T] \quad (11)$$

Here we have used a literature value of 1.11 for the Gruneisen parameter, γ (Liu and Li, 2006). Fitting the K_S at high pressures and 300 K using the third finite equation with a fixed $K_{S0}=129.8(9)$ GPa and a self-consistent density model (Speziale and Duffy, 2002), we have obtained $K_{S0}'=4.45(5)$ (Figs. 4 and 5). The obtained K_{S0}' is slightly greater than that in the literature (Abramson et al., 1997; Zha et al., 1998). With the determined K_{S0}' value and high P-T K_S results, the temperature derivative of the bulk modulus, $(\partial K_S / \partial T)_P$, is determined to be -0.020(2) GPa/K. The derived thermal expansion coefficient at ambient conditions is $3.47 \times 10^{-5} \text{ K}^{-1}$.

As noted above, there is an enhanced effect of temperature on C_{12} and C_{13} towards higher pressures (Fig. 3). An extra term in the second pressure derivative of the C_{12} and C_{13} is needed in our modeling in order to fully describe the enhanced temperature dependence at higher pressures: $C_{12}'' = -0.015 - 0.00021 \times T$ (GPa/K), and $C_{13}'' = -0.015 - 0.00021 \times T$ (GPa/K). For C_{11} , the pressure derivative is derived from the high P-T data. The P-T derivatives of the shear modulus, G , are evaluated using the following equations:

$$G_0(T) = G_0(300) + (\partial G / \partial T)_P (T - 300) \quad (12)$$

$$G = (1 + 2f)^{5/2} [G_0(T) + b_1 f + 1/2 b_2 f^2] \quad (13)$$

$$b_1 = 3K_{T0}(T)G'_0 - 5G_0(T) \quad (14)$$

265
$$b_2 = 9\{K_{T_0}^2(T)[G''_0 + 1/K_{T_0}(T)(K'_{T_0} - 4)G'_0] + 35G_0(T)/9\} \quad (15)$$

266 We thus obtain: $G'_0 = 1.80(3)$, $G''_0 = -0.10(3)$ GPa⁻¹ and $(\partial G/\partial T)_P = -0.010(2)$ GPa/K.

267

268 **4. Discussion and Geophysical Implications**

269 **4.1. Single-crystal elasticity of olivine at high P-T**

270 We have derived all of the nine elastic constants of single-crystal San Carlos olivine
271 (Fig. 3 and Table S1). At ambient conditions, all of the elastic moduli are in excellent
272 agreement with results in literature (Abramson et al., 1997; Webb, 1989; Zha et al.,
273 1998). At 300 K, the longitudinal, off-diagonal and aggregate bulk moduli at high
274 pressures are consistent with results by Abramson et al. (1997) and Webb (1989)
275 within experimental uncertainties. Moreover, we have observed a softening in the
276 shear moduli, C_{44} , C_{55} , C_{66} and G , with increasing pressure when approaching the
277 olivine to wadsleyite phase transition (Fig. 3). Similar softening of elastic constants
278 with pressure due to a structural phase transition has also been shown in other
279 minerals, including stishovite, enstatite and orthoferrosilite (Kung and Li, 2014; Li et
280 al., 2014; Yang and Wu, 2014). The softening of elastic constants, however, is not
281 observed by Webb (1989) likely because of the limited experimental pressure range
282 (up to 3 GPa). Abramson et al. (1997) reported the softening in C_{55} . However, other
283 shear moduli, C_{44} , C_{66} and G , in Abramson et al. (1997) were only determined up to
284 12 GPa, where the softening started to occur. Although similar softening in the shear
285 moduli has been shown by Zha et al. (1998) using the same Brillouin technique, C_{55}
286 and C_{66} in this study exhibit a stronger downward curvature than those in Zha et al.

287 (1998) (Fig. 3). Moreover, C_{11} and C_{13} at high pressures in Zha et al. (1998) are much
288 lower than our results. To independently determine all elastic constants of minerals in
289 the orthorhombic system, previous studies have showed that measuring velocities
290 using platelets close to basal planes, such as (100), (010), and (001), permits reliable
291 derivations of all elastic constants (Jiang et al., 2004, 2009; Speziale et al., 2004;
292 Wang et al., 2014), because some of the longitudinal and/or shear moduli in such
293 planes are not covariance with other elastic constants and can be better constrained.
294 Even so, it is still necessary to measure the velocity in two or three different crystal
295 planes, because single-crystal elastic moduli of a given orthorhombic mineral may not
296 be well constrained when lacking sufficient velocity measurements (Every, 1980; Fan
297 et al., 2015). It should be noted that the elastic moduli in Zha et al. (1998) were
298 retrieved from the velocity data in the (111) plane with 19 measurements at each
299 given pressure such that some of the obtained C_{ij} s in Zha et al., (1998) are expected to
300 suffer from a stronger tradeoff with each other because these C_{ij} s cannot be
301 well-constrained. Therefore, we have conducted our measurements using two platelets
302 in different crystallographic orientations in which one platelet is close to the basal
303 plane while another is a random plane in order to better constrain all nine elastic
304 constants for the orthorhombic olivine crystal.

305

306 We have further compared our elasticity results to literature values for San Carlos
307 olivine at high temperatures (Figs. 3 and 4, Table S2) (Isaak, 1992). Extrapolating our
308 elastic constants at high P-T back to 1 bar and high temperature demonstrates

309 agreement with Isaak (1992), with the exception of the effect of temperature on C_{44}
310 and C_{23} . We note that the temperature derivatives at 1 bar and high temperatures for
311 some of the elastic moduli (including C_{22} , C_{33} , C_{44} , C_{55} , and C_{66}) are indistinguishable
312 from the values obtained from the high P-T measurements within experimental
313 uncertainties (Table S2) (Isaak, 1992). However, the temperature dependence of C_{11}
314 derived after considering the combined effect of P-T is smaller than the one at 1 bar
315 and high temperatures, whereas the temperature dependence of C_{23} is larger. Of
316 particular importance is the enhanced temperature dependence of the off-diagonal
317 moduli, C_{12} and C_{13} , at high pressures. The softening behavior observed at high P-T
318 may be associated with the olivine-wadsleyite transition that is prohibited because of
319 the kinetic energy barrier (Kung and Li, 2014; Li et al., 2014; Yang and Wu, 2014).
320 We have also plotted K_S and G as a function of temperature at various pressures to
321 show the effect of temperature on the elasticity of olivine at a given pressure (Fig. 5).
322 At the investigated P-T range, K_S and G exhibit a nearly linear decrease with
323 increasing temperature at a given pressure.

324

325 4.2. The velocity contrast at the 410-km depth

326 The seismic 410-km discontinuity is associated with a 4-5% increase in V_P and a
327 4-4.6% increase in V_S , and it has been widely accepted to be caused by the olivine to
328 wadsleyite phase transition (Grand and Helmberger, 1984; Kennett et al., 1995;
329 Lefevre, 1989; Walck, 1985). A long-standing problem in mineral physics is that the
330 calculated V_P and V_S jumps in a pyrolite composition using literature elasticity results

331 of olivine and wadsleyite are 6-8% after accounting for their relative proportions.
332 These values are greater than the aforementioned seismic discontinuities of 4-5% at
333 the 410-km depth (Cammarano et al., 2005b; Duffy and Anderson, 1989; Li and
334 Liebermann, 2007; Li et al., 2001). To understand the difference between the pyrolite
335 model and seismic observations at the 410-km depth, we have constructed a new
336 pyrolitic velocity model for the upper mantle and transition zone using our elasticity
337 of olivine at high P-T (Fig. 6).

338

339 In the pyrolite compositional model, the volume percentage of olivine is
340 approximately 60% (Ringwood, 1975), while pyroxene and garnet, so-called basaltic
341 components, account for another 40% of the system (Irifune, 1987; Jeanloz and
342 Thompson, 1983). Olivine transforms into wadsleyite at approximately 410-km depth
343 and to ringwoodite at around 520-km depth, whereas pyroxene and garnet transform
344 into majorite in the upper mantle and transition zone (Fei and Bertka, 1999). In our
345 modeling, we have taken into account the phase relations as a function of depth as
346 well as the partitioning of Fe between olivine polymorphs and between olivine and
347 residual basaltic component at the phase equilibria using previous experimental and
348 theoretical results (Akaogi et al., 1989; Angel et al., 1992; Irifune, 1987; Irifune and
349 Isshiki, 1998; Irifune and Ringwood, 1987; Katsura and Ito, 1989; Pacalo and
350 Gasparik, 1990; Stixrude and Lithgow-Bertelloni, 2005; Xu et al., 2008). After
351 considering the variation in phases and compositions with depth, the velocity of
352 pyrolite has been constructed using the elasticity of olivine in this study and elasticity

353 of other mantle phases in previous studies (Table 1) (Chai et al., 1997; Finger and
354 Ohashi, 1976; Isaak et al., 2010; Jackson et al., 2003; Jackson et al., 2007; Jiang et al.,
355 2004b; Lu et al., 2013; Sang and Bass, 2014; Sinogeikin and Bass, 2000, 2002;
356 Sinogeikin et al., 2001; Sinogeikin et al., 1998; Wang et al., 2014; Zhao et al., 1997;
357 Zou et al., 2012).

358

359 The computed velocity model for a pyrolitic composition is compared to literature
360 seismic profiles (Fig. 6). In general, the modeled V_P profile of the system is consistent
361 with seismic observations in the upper mantle, whereas V_S is slightly larger. Karato
362 (1993) has pointed out that the anelastic effects should be considered when applying
363 the experimental data in interpreting the seismic velocities of the mantle. If we
364 consider the anelastic effects due to temperature, the modeled V_S profile can decrease
365 by 0.6-1.4%, leading to a V_S of the modeled pyrolitic mantle in a better agreement
366 with seismic observations (Cammarano et al., 2003). However, due to the discrepancy
367 in the attenuation profile of the upper mantle and transition zone in seismic studies
368 (e.g. Romanowicz and Durek, 2000), the effect of anelasticity is not included in our
369 modeling. On the other hand, a previous experimental study has shown that V_S of
370 mantle minerals, such as majorite, could suffer from an enhanced non-linear reduction
371 with increasing temperature at high pressures (Irifune et al., 2008). Taking this
372 non-linear temperature effect at high pressures into account could lower the modeled
373 V_P and V_S profiles that are in a better agreement with the seismic velocity profiles
374 (Irifune et al., 2008). Although the elasticity of Fe-bearing olivine has been found to

375 decrease linearly with temperature in both this study and previous experiments
376 performed up to 1500 K and 1 bar (Isaak, 1992), it remains unclear whether or not the
377 elasticity and sound velocities of olivine will suffer from a non-linear decrease with
378 temperature above 900 K at high pressures. The aforementioned high-temperature
379 effects on the attenuation and nonlinear velocity decrease should be further
380 investigated in future studies in order to add to our understanding of the seismic
381 features and mineralogical models of the region.

382

383 For the 410-km discontinuity, the olivine to wadsleyite phase transition produces a
384 sharp velocity jump with a width of ~ 7 km (Fig. 6) (Frost, 2003). The calculated
385 velocity jumps at the 410-km depth are 6.4% for V_P and 6% for V_S . Although the
386 velocity jumps shown here are in a much better agreement with seismic observations
387 as compared to previous velocity profiles for a pyrolitic upper mantle (Cammarano et
388 al., 2005b; Duffy and Anderson, 1989; Li and Liebermann, 2007; Li et al., 2001),
389 these values are still greater than the seismically-observed velocity increase of 4-5 %
390 at the 410-km depth (Grand and Helmberger, 1984; Kennett et al., 1995; Lefevre,
391 1989; Walck, 1985). To match a 4-5% velocity jump in seismic observations of the
392 region, the olivine content in the mantle is estimated to be ~ 40 -50 vol%, a value that
393 is lower than what is expected for a pyrolitic compositional model; though, future
394 elasticity studies at compatible P-T conditions of the mantle with higher accuracy are
395 needed to better address this issue. On the other hand, the presence of water in the
396 upper mantle and transition zone may help to reconcile some of the differences

397 between the pyrolite model and seismic images (Jacobsen et al., 2008; Mao et al.,
398 2011; Mao et al., 2008a; Mao et al., 2008b). Recent geological, mineralogical, and
399 seismic studies have shown that various locations of the upper mantle and transition
400 zone could be hydrated with up to 1 wt.% H₂O in ringwoodite (Pearson et al., 2014;
401 Schmandt et al., 2014; Song et al., 2004; van der Meijde et al., 2003). Since hydration
402 can cause a greater reduction in the velocity of wadsleyite than olivine at mantle P-T
403 conditions, the presence of a certain amount of water in the mantle will lower the
404 velocity jump at the 410-km depth (Jacobsen et al., 2008; Mao et al., 2011; Mao et al.,
405 2008a; Mao et al., 2010; Mao et al., 2008b), therefore making the velocity of pyrolite
406 agree better with seismic observations of the upper mantle and the transition zone
407 (Mao et al., 2011; Mao et al., 2008a; Mao et al., 2008b). Future studies are needed to
408 further examine the effect of water on the velocity of minerals in a pyrolitic
409 composition in the upper mantle and transition zone.

410

411 4.3. Velocity profiles of the metastable olivine wedge in the subduction slabs

412 The metastable olivine wedge in the subducted mantle lithosphere has long been
413 proposed to exist below the 410-km discontinuity due to the relatively cold nature of
414 the slabs that kinetically inhibits the transformation of olivine to wadsleyite in the
415 region. According to previous petrological studies, subducted mantle lithosphere
416 consists of harzburgite at the top and less depleted lherzolite at the bottom (Irifune
417 and Ringwood, 1993; Poli and Schmidt, 2002; Stern, 2002). The volume percentage
418 of olivine is typically greater than 60% in harzburgite and is approximately 40% in

419 lherzolite (Hacker et al., 2003; Poli and Schmidt, 2002; Song et al., 2009; Stern et al.,
420 2012). The relatively low temperatures in the cold slabs will kinetically inhibit the
421 phase transition of olivine to wadsleyite at the depth of 410-km, such that olivine in
422 the slabs likely transforms to ringwoodite directly at a greater depth in the transition
423 zone, potentially leading to the occurrence of deep earthquakes at greater than 410-km
424 depth (Chen et al., 2004; Green and Burnley, 1989; Green et al., 1990; Kirby, 1987;
425 Kirby et al., 1996; Mosenfelder et al., 2001; Raterron et al., 2002). If the phase
426 transition of metastable olivine is the major cause for deep earthquakes, the answer to
427 deep earthquakes resides in the seismic signatures and detectability of the metastable
428 olivine wedge. Recently, a number of seismic studies have reported the existence of a
429 low-velocity and anisotropic wedge with 3-5% lower velocities and shear-wave
430 splitting of ~ 1 s in the slabs below the 410-km depth, which was viewed as evidence
431 for the presence of the metastable olivine in the subducted slabs (e.g. Jiang et al., 2008;
432 Kaneshima et al., 2007; Liu et al., 2008; Pankow et al., 2002; Tono et al., 2009). It is
433 thus of great interest to model the velocity of metastable olivine in the transition zone
434 depth. Comparing the modeled velocity results to seismic observations will provide
435 crucial mineralogical constraints on the seismic signatures of metastable olivine.

436

437 Using our new high P-T elasticity results, we have modeled the velocities and
438 anisotropies of the metastable olivine in the cold slabs. Previous studies have shown
439 that the kinetic cut-off temperature for the existence of metastable olivine down to a
440 depth of 600 km is ~ 873 - 973 K which is similar to our maximum experimental

temperatures (Liu, 1983; Sung and Burns, 1976). Here we assume that the core of the
slabs is at 900 K at the transition zone for the computation of the sound velocities of
olivine (Fig. 7). Above the 410-km depth, metastable olivine at 900 K has a V_P and V_S
~4% greater than that of an expected normal mantle geotherm. Once the slab crosses
the 410-km discontinuity, the V_P and V_S of olivine in the slabs will be ~6% lower than
that of wadsleyite in the normal mantle between 410 and 520-km depth and ~8-11%
lower than that of ringwoodite between 520 and 660-km depth. Assuming that the
olivine content is 60 vol% in the mantle lithosphere as well as in the normal mantle,
the velocity in the core of slabs will be ~3.6% lower than that in the normal mantle at
410 to 520-km depth and ~4.8-6.6% lower than that at 520 to 660-km depth. In this
case, the presence of metastable olivine in the slabs will produce a seismic signature
with lower velocities than the surrounding mantle materials, consistent with a number
of seismic observations below the 410-km depth (Iidaka and Suetsugu, 1992; Jiang et
al., 2008; Kaneshima et al., 2007).

In addition to the low-velocity wedge in the slabs, seismic anisotropies have also been
identified below the 410-km depth (Liu et al., 2008; Sandvol and Ni, 1997; Tono et al.,
2009). The observed seismic anisotropy in the slabs in the mantle transition zone
could be caused by the preferred alignment of the metastable olivine crystals (Liu et
al., 2008; Mainprice, 2007; Sandvol and Ni, 1997; Tono et al., 2009). Using the
obtained single-crystal elasticity, we have computed the maximum azimuthal
compressional-wave anisotropy, $A_P [(V_{P,max}-V_{P,min})/V_{P,ave}]$, and shear-wave splitting,

463 $A_S^{\text{PO}} [(V_{S1}-V_{S2})/V_{S,\text{aggr}}]$, of olivine at various slab temperatures (Fig. 8). In general,
464 olivine has a much greater A_P and A_S^{PO} than that of wadsleyite and ringwoodite along
465 an expected mantle geotherm (Fig. 8). Decreasing temperature only slightly lowers
466 the A_P of olivine. Even if the temperature in the core of slabs is as low as 873 K, the
467 A_P of the metastable olivine in the transition zone is still twice as much as A_P of
468 wadsleyite in the normal mantle and is seven times greater than the A_P of ringwoodite.
469 Meanwhile, we have also noted that A_P calculated using our elasticity of olivine
470 considering the combined P-T effect is greater than those results obtained using
471 literature results (Issak, 1992; Zha et al., 1998), showing the importance of
472 investigating the elasticity of mantle minerals at simultaneously high P-T conditions
473 relevant to the region.

474

475 In contrast to A_P , A_S^{PO} of olivine exhibits a non-linear variation with depth caused by
476 the softening of the shear moduli and the enhanced temperature dependence on the
477 off-diagonal moduli at high pressures (Fig. 8). In particular, A_S^{PO} of olivine along an
478 expected mantle geotherm decreases with depth up to 290-km but starts to increase at
479 greater depths. Such abnormal increase in A_S^{PO} at a greater depth cannot be
480 recognized without the knowledge of the combined P-T effect on the elasticity of
481 single-crystal olivine (Fig 8). In the cold slabs with a temperature of 873 K where the
482 metastable olivine maybe present, A_S^{PO} will keep decreasing with increasing depth up
483 to the 410-km discontinuity, but it will become more enhanced in the transition zone
484 region. Along the mantle geotherm, A_S^{PO} of olivine is much greater than that of

485 wadsleyite and ringwoodite. According to our modeled results, the metastable olivine
486 in the cold slabs with a temperature of 873-1273 K has an A_S^{PO} similar to that of
487 wadsleyite in the normal mantle between 410 and 520-km depth. From 520-km depth
488 to the bottom of the transition zone, A_S^{PO} of the metastable olivine is still significantly
489 greater than that of ringwoodite.

490

491 Based on the aforementioned discussions, the presence of metastable olivine in the
492 subduction slabs can be used to help explain the observed low-velocity layers and
493 seismic anisotropies in the region. We need to point out that both A_P and A_S^{PO} shown
494 here represent the maximum velocity anisotropies of single-crystal olivine at mantle
495 P-T conditions. Yet modeling the anisotropy of the mantle requires detailed
496 knowledge of the evolution of the lattice preferred orientation (LPO) of
497 polycrystalline olivine with increasing P-T together with our obtained elastic moduli
498 of single-crystal olivine (Mainprice, 2007; Mainprice et al., 2000). However, previous
499 deformation experiments, which provide crucial constraints on the LPO and slip
500 systems of olivine, have been rarely performed at transition zone pressures (e.g.
501 Demouchy et al., 2009; Durham et al., 1977; Faul et al., 2011; Hilairt et al., 2012;
502 Jung and Karato, 2001; Katayama et al., 2004). Even the low pressure and high
503 temperature experiments yield conflicting results (e.g. Demouchy et al., 2009;
504 Durham et al., 2009; Kawazoe et al., 2009; Li et al., 2006). In this case, modeling the
505 anisotropy of subduction slabs using future deformation results for the LPO of olivine
506 at relevant P-T conditions of the transition zone and our elastic moduli of olivine will

507 provide more comprehensive and reliable constraints on the anisotropy of metastable
508 olivine in the slabs.

509

510 In summary, the full set of elastic constants of single-crystal olivine has been
511 measured for the first time using combined Brillouin spectroscopy and X-ray
512 diffraction at high P-T conditions. These results provide tight experimental constraints
513 on the combined P-T effects on the sound velocity profiles and seismic anisotropies of
514 olivine in the upper mantle. Although most of the elastic moduli increase with
515 increasing pressure at given high temperatures, three moduli, C_{11} , C_{12} and C_{13} exhibit
516 stronger temperature-induced reduction at greater pressures, showing the importance
517 of the combined effects of high P-T on the elasticity of mantle minerals. The elasticity
518 of single-crystal olivine at high P-T is modeled and applied to decipher the seismic
519 discontinuity at the 410-km depth in a pyrolitic mantle composition as well as the
520 velocity profiles of the metastable olivine wedge in the subduction slabs. Considering
521 the combined high P-T effects on the elasticity of olivine along an expected mantle
522 geotherm, the velocity jumps at the 410-km depth are 6.0% for V_P and 5.3% for V_S ,
523 which are more consistent with seismic observations, although other factors such as
524 the addition of water or compositional effects may still be needed to reconcile the
525 difference between current mineral physics results and seismic observations. We also
526 found that the presence of the metastable olivine in the slabs can exhibit both low
527 velocities and strong seismic anisotropies that are consistent with seismic
528 observations of the region and will help to locate the potential metastable olivine

wedge in the slabs. Our in situ high P-T results, together with thermoelastic modeling, provide new insights on the composition and seismic structures of the Earth's mantle.

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891

Table 1. Elasticity of minerals in the upper mantle and transition zone

Mineral	ρ (g/cm ³)	K_{S0} (GPa)	K_{S0}'	$\partial K_S / \partial T$ (GPa/K)	$\partial^2 K_S / \partial T^2$ (GPa/K ²)	G (GPa)	G_0'	G'' (1/GPa)	$\partial G / \partial T$ (GPa/K)	$\partial^2 G / \partial T^2$ (GPa/K ²)
olivine ^a	3.227+0.012X _{Fe} ⁱ	128.8+0.08X _{Fe}	4.47	-0.02		81.6-0.38X _{Fe}	1.8	-0.1	-0.01	
wadsleyite ^b	3.474+0.0159X _{Fe}	170	4.2	-0.171		112-0.4X _{Fe}	1.4		-0.0157	
ringwoodite ^c	3.559+0.014X _{Fe}	184	4.1	-0.021		120-0.25X _{Fe}	1.4		-0.016	
clinopyroxene ^d	3.327	117.2	4.2	-0.013		72.2	1.4		-0.01	
orthopyroxene ^e	3.306	115.5	7.82	-0.0238		78.1	1.45		-0.0125	
HP clinopyroxene ^f	3.45	156.7	5.5	-0.017		98.5	1.5		-0.015	
pyrope garnet ^g	3.567	171.2	4.1	-0.0168		93.7	1.3		-0.0051	
majorited- garnet ^h	3.605	164.4	4.2	-0.0091	-6×10 ⁻⁶	94.9	1.1		-0.0074	-5×10 ⁻⁶

^aThis study; Abramson et al. (1997);Zha et al. (1996); ^bIsaak et al. (2007); Sinogeikin et al. (1998a); Wang et al. (2014);Zha et al. (1997); ^cSinogeikin et al. (1998a); Sinogeikin et al. (2003); ^dCollins and Brown (1998);Duffy and Anderson (1989); ^eChai et al. (1997); Jackson et al. (2007); $K_S=115.4+7.82P-0.18P^2$; ^f Kung et al. (2005); ^gSinogeikin and Bass (2000); Lu et al. (2013); ^hIrifune et al. (2008); ⁱ Fe content in mole.%

906 **Figure caption**

907 Figure 1. Representative Brillouin spectrum of olivine at 13.3 GPa and 900 K. Black
908 line: collected data; red, blue, green and orange lines: fitting results. Insert figure is a
909 representative sample photo taken at ~6 GPa and 300 K.

910

911 Figure 2. Representative acoustic velocities of olivine measured at high P-T
912 conditions as a function of azimuthal angles. Circles: measured velocities; lines:
913 fitting results; black: quasi-longitudinal modes; red: fast quasi-shear mode; blue: slow
914 quasi-shear mode.

915

916 Figure 3. Elastic moduli of single-crystal San Carlos olivine at high P-T. Blue: at 300
917 K; green: at 500 K; orange: at 750 K; red: at 900 K; solid circles: this study; open
918 circles: Isaak (1992); dashed line: Abramson et al. (1997); dotted line: Zha et al.
919 (1998); dashed-dotted line: Webb (1989).

920

921 Figure 4. Aggregate elastic moduli of San Carlos olivine at high pressures and
922 temperatures. Blue: at 300 K; green: at 500 K; orange: at 750 K; red: at 900 K; solid
923 circles: this study; open circles: Isaak (1992).

924

925 Figure 5. Aggregate elastic moduli of San Carlos olivine. (a). At 300 K. Literature
926 results are also plotted for comparison. Blue: this study; black: Zha et al. (1998);
927 orange: Abramson et al. (1997); green: Webb (1989). (b). Calculated with increasing

928 temperature at a given pressure.

929

930 Figure 6. Compressional (V_P) and shear wave (V_S) velocities of the mantle in a
 931 pyrolite composition compared with seismic profiles. Red line: calculated velocities
 932 of mantle in a pyrolite composition; black line: AK135 (Kennett et al., 1995); blue
 933 line of V_P : CJF (Walck, 1985); green line of V_P : S25 (Lefevre, 1989); blue line of V_S :
 934 SNA (Grand and Helmberger, 1984); green line of V_S : TNA (Grand and Helmberger,
 935 1984). For modeling the elasticity of wadsleyite at relevant high pressure-temperature
 936 conditions of the transition zone, the effect of high pressure is taken from Wang et al.
 937 (2014), while the effect of temperature is from Isaak et al. (2010).

938

939 Figure 7. Compressional (V_P) and shear wave (V_S) velocities of olivine polymorphs in
 940 the Earth's mantle. (a). Blue lines: velocities of olivine at 900 K, representing the
 941 velocities of olivine in the cold subduction slabs; red lines: velocities of olivine,
 942 wadsleyite and ringwoodite along an expected mantle geotherm (Brown and
 943 Shankland, 1981); (b). Velocity difference between olivine at 900 K in the slabs and
 944 olivine polymorphs along an expected mantle geotherm (Brown and Shankland, 1981).
 945 Black line: V_P ; red line: V_S .

946

947 Figure 8. Velocity anisotropy of olivine at different temperatures in the slab compared
 948 with olivine polymorphs along an expected mantle geotherm. (a). Maximum
 949 compressional wave anisotropy, A_P . (b). Maximum shear wave splitting, A_S^{PO} . Red,

950 orange, green, blue and black lines: olivine at 1200, 1000, 800, and 600°C in the slabs,
951 respectively; wine lines: olivine, wadsleyite and ringwoodite along the mantle
952 geotherm (Brown and Shankland, 1981); dark yellow: calculated using the literature
953 high pressure, 300 K and high temperature, 1 bar results of olivine along the mantle
954 geotherm for comparison (Issak, 1992; Zha et al., 1998).

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