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# Sound velocities and single-crystal elasticity of hydrous Fo90 olivine to 12 GPa

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## Abstract

Nominally anhydrous minerals (NAMs) may contain significant amounts of water and constitute an important reservoir for mantle hydrogen. The colloquial term ‘water’ in NAMs is related to the presence of hydroxyl-bearing (OH<sup>-</sup>) point defects in their crystal structure, where hydrogen is bonded to lattice oxygen and is charge-balanced by cation vacancies. This hydrous component may therefore have substantial effects on the thermoelastic parameters of NAMs, comparable to other major crystal-chemical substitutions (e.g., Fe, Al). Assessment of water concentrations in natural minerals from mantle xenoliths indicates that olivine commonly stores ~10<sup>0</sup> – 10<sup>2</sup> ppm of water. However, the lack of samples originating from depths exceeding ~250 km coupled with the rapid diffusion of hydrogen in olivine at magmatic temperatures makes the determination of the olivine water content in the upper mantle challenging. On the other hand, numerous experimental data show that, at pressures and

temperatures corresponding to deep upper mantle conditions, the water storage capacity of olivine increases to 0.2 – 0.5 wt.% H<sub>2</sub>O. Therefore, determining the elastic properties of olivine samples with more realistic water contents for deep upper mantle conditions may help in interpreting both seismic velocity anomalies in potentially hydrous regions of Earth's mantle as well as the observed seismic velocity and density contrasts across the 410-km discontinuity.

Here, we report simultaneous single-crystal X-ray diffraction and Brillouin scattering experiments at room temperature up to 11.96(2) GPa on hydrous (0.20(3) wt.% H<sub>2</sub>O) Fo90 olivine to assess its full elastic tensor. To place further constraints on the effect of hydration on olivine elastic properties, we modelled higher water concentrations in olivine, namely 0.5 wt.% H<sub>2</sub>O, using our new accurate data. Although the elastic moduli and pressure derivatives of hydrous Fo90 olivine are slightly different compared to those of the corresponding anhydrous phase, our results demonstrate that the sound wave velocities of hydrous and anhydrous olivines are indistinguishable within uncertainties at pressures corresponding to the base of the upper mantle. Contrary to previous claims, our data suggest that water in olivine is not seismically detectable, at least for contents consistent with deep upper mantle conditions. In addition to that, our data reveal that the hydration of olivine is unlikely to be a key factor in reconciling seismic velocity and density contrasts across the 410-km discontinuity with a pyrolitic mantle.

**Keywords:** hydrous olivine, Brillouin scattering, elasticity, high pressure; NAMs

## 1. Introduction

Although commonly referred to as nominally anhydrous minerals (NAMs), the dominant phases of Earth's upper mantle may contain significant amounts of water (e.g., Bell and Rossman, 1992). The occurrence of water in NAMs is closely related to the presence of hydrogen in their crystal structures, which is bonded to oxygen atoms forming hydroxyl-bearing (OH<sup>-</sup>) point defects and its incorporation is typically charge-balanced by the formation of cation vacancies. Olivine,  $\alpha$ -(Mg,Fe)<sub>2</sub>SiO<sub>4</sub>, is considered to be the most abundant mineral of Earth's upper mantle and to constitute

52 about 60 vol.% of pyrolitic phase assemblages (Ringwood, 1975). The water concentration detected  
53 in natural olivine samples from mantle xenoliths is generally low, in the order of  $10^0 - 10^2$  ppm wt.  
54 (Beran and Libowitzky, 2006; Novella et al., 2015; Peslier et al., 2010). Although a growing body of  
55 water analyses of olivine is available in the literature (Bonadiman et al., 2009; Peslier, 2010; Xia et  
56 al., 2010, and many other reviews), trends between water content and its distribution across different  
57 geological settings are still difficult to determine, as well as the actual abundance of water in olivine  
58 in the upper mantle. This is due to both the rapid diffusion of hydrogen in olivine at magmatic  
59 temperatures (e.g., Demouchy and Mackwell, 2006), which leads to partial dehydration of olivine in  
60 mantle xenoliths during their ascent, and the lack of xenoliths originating from depths greater than  
61 ~250 km. Even though the fast ascent of kimberlite magmas should prevent dehydration in olivine  
62 during xenolith transport (Demouchy et al., 2006; Peslier et al., 2008), in contrast to olivines from  
63 xenoliths found in alkali basalts which are commonly affected by H loss (e.g., Peslier et al., 2008),  
64 our understanding of how the water content of olivines changes as a function of depth largely relies  
65 on experimental studies. Indeed, a plethora of experimental data on water solubility in olivines at  
66 pressures and temperatures relevant to the upper mantle are available and indicate much higher water  
67 contents compared to those detected in natural olivine samples, especially at deep upper mantle  
68 conditions where the water storage capacity of olivine increases to 0.2 – 0.5 wt.% H<sub>2</sub>O (e.g., Férot  
69 and Bolfan-Casanova, 2012; Hirschmann et al., 2005; Mosenfelder, 2006). Therefore, synthetic  
70 olivine samples with a more realistic water content expected for deep upper mantle conditions warrant  
71 investigation, as this may improve our understanding of various geodynamic processes operating on  
72 Earth (Regenauer-Lieb, 2006, and references therein).

73 Water incorporation in olivine, even in trace amounts, has long been known to have substantial  
74 effects on its physical and chemical properties, such as atomic diffusivity, electrical conductivity,  
75 thermal conductivity, rheology, and melting (e.g., Chang et al., 2017; Costa and Chakraborty, 2008;  
76 Inoue, 1994; Jung and Karato, 2001; Smyth et al., 2006). From a crystal-chemical perspective,  
77 hydrogen incorporation in the crystal structure of olivine may occur both in the octahedral M sites,

78  $\text{Mg}^{2+} \leftrightarrow 2\text{H}^+$ , and in tetrahedral T sites,  $\text{Si}^{4+} \leftrightarrow 4\text{H}^+$  (e.g., Brodholt and Refson, 2000; Hushur et al.,  
79 2009; Smyth et al., 2006). Previous experimental studies about the substitution mechanism of water  
80 in olivine showed contrasting results, although it was recognized that the synthesis conditions  
81 (pressure and  $\text{SiO}_2$  activity) were influencing factors (e.g., Bali et al., 2008; Férot and Bolfan-  
82 Casanova, 2012; Matveev et al., 2001; Withers and Hirschmann, 2008). However, recent electron  
83 microprobe (EPMA) data coupled with transmission Fourier transform infrared spectroscopy (FTIR)  
84 analyses (Fei and Katsura, 2020), solid-state nuclear magnetic resonance (NMR) spectroscopy (Xue  
85 et al., 2017), and first-principles calculations studies (Umemoto et al., 2011; Xue et al., 2017) suggest  
86 that hydrogen substitution in olivine mainly occurs at the T sites at high pressure. Therefore, hydrogen  
87 incorporation in the M sites can be ruled out from being the predominant substitution mechanism in  
88 olivine under typical mantle conditions, as its occurrence is restricted to shallow depths ( $\leq 2.5$  GPa)  
89 and high  $\text{SiO}_2$  activity (Bali et al., 2008; Fei and Katsura, 2020; Withers et al., 2011).

90 Despite its possible geophysical relevance, the effect of water on the elastic properties of olivine  
91 has been poorly constrained. The only elasticity measurements available in the literature were  
92 conducted by Mao et al. (2010) on hydrous (0.9 wt.%  $\text{H}_2\text{O}$ ) end-member forsterite (Fo100) single  
93 crystals up to 14 GPa and by Jacobsen et al. (2008, 2009) on hydrous (0.8 wt.%  $\text{H}_2\text{O}$ ) Fo97 olivine  
94 single crystals at ambient pressure. These works showed that the incorporation of water into pure  
95 forsterite and near end-member compositions is accompanied by a considerable reduction of the bulk  
96 ( $K_S$ ) and shear ( $G$ ) moduli at ambient conditions. However, owing to the much larger moduli pressure  
97 derivatives of the hydrated samples,  $K_S$  and  $G$  of the hydrous Fo100 at high pressure are greater than  
98 those of the corresponding anhydrous phase (Mao et al., 2010). Nonetheless, the effect of hydration  
99 on the elasticity of the more relevant Fo90 olivine mantle composition remains largely unconstrained  
100 because no experimental data are available. Furthermore, previous works studied hydrous samples  
101 containing 0.8-0.9 wt.%  $\text{H}_2\text{O}$ , which is well above the expected water content for olivine at deep  
102 upper mantle conditions (e.g., Férot and Bolfan-Casanova, 2012; Hirschmann et al., 2005;  
103 Mosenfelder, 2006).

104 In this work, we set out to investigate the effect of hydration on the elastic properties and sound  
105 wave velocities of hydrous Fo90 olivine samples with more realistic water content for deep upper  
106 mantle conditions, as it may help in interpreting both seismic velocity anomalies in potentially  
107 hydrous regions of Earth's mantle as well as the observed seismic velocity and density contrasts across  
108 the 410-km discontinuity. To this aim, we performed simultaneous single-crystal X-ray diffraction  
109 (SCXRD) and Brillouin scattering measurements at room temperature up to 11.96(2) GPa on hydrous  
110 (0.20(3) wt.% H<sub>2</sub>O) Fo90 olivine to constrain its full elastic tensor. By comparing our new accurate  
111 data with those available from literature for anhydrous Fo90 olivine, we evaluated the combined  
112 effect of H<sub>2</sub>O and Fe incorporation on the pressure-dependent elasticity of olivine. Our measurements  
113 are suitable to determine the sound wave velocities of hydrous Fo90 olivine at pressures  
114 corresponding to the base of the upper mantle, to be integrated in theoretical/experimental studies  
115 attempting to constrain the olivine abundance and water content at the 410-km discontinuity.

## 116 **2. Materials and Methods**

117 Large (200 – 500  $\mu$ m) and homogeneous single crystals of hydrous Fo90 olivine used in this study  
118 were previously synthesized and characterized by Withers et al. (2011, 2012, sample M475). Major  
119 element concentrations determined by wavelength-dispersive EPMA analysis indicate the Mg# of  
120 synthesized olivines to be 90.2(1), whereas their water content was determined to be ~0.20(3) wt.%  
121 based on elastic recoil detection analysis (ERDA) (Withers et al., 2012). Polarized FTIR spectroscopy  
122 and secondary ion mass spectroscopy (SIMS) measurements were also conducted on the same  
123 sample, showing excellent agreement with one another (Withers et al., 2012).

124 High-pressure Brillouin scattering measurements in DAC are conducted in the so-called platelet  
125 geometry (Whitfield et al., 1976). For olivine, at least two crystal platelets with different  
126 crystallographic orientations are required to obtain the nine independent elastic stiffness coefficients  
127 ( $c_{ij}$ ) given its orthorhombic symmetry. The full elastic tensor is obtained by a least-square fit of  
128 experimentally determined densities and sound wave velocities measured in different crystallographic  
129 directions using the Christoffel's equation:

$$|c_{ijkl}q_jq_l - \rho v_i^2 \delta_{ik}| = 0 \quad (1)$$

where  $c_{ijkl}$  are the elastic stiffness coefficients in tensorial notation (e.g., Nye, 1985),  $q_j$  and  $q_l$  are direction cosines,  $\rho$  is the density,  $v_i$  are the sound wave velocities and  $\delta_{ik}$  is the Kronecker delta. To obtain accurate and precise  $c_{ij}$  values, it is important that their correlation in the fit procedure is low. This is achieved by using two appropriate crystallographic orientations (Criniti et al., 2021). We used published  $c_{ij}$  data for Fo90 olivine (Abramson et al., 1997) to simulate the shear ( $v_s$ ) and compressional velocities ( $v_p$ ) for different crystallographic planes. Synthetic  $v_s$  and  $v_p$  were randomly scattered by up to  $\pm 30$  or  $\pm 60$  m/s, respectively, to simulate realistic datasets and to obtain realistic uncertainties in the inversion procedure. Platelets with direction cosines (0.32, 0.91, 0.26) and (0.78, 0.02, 0.62), corresponding to 101 and 161 *hkl* indices, were chosen for high-pressure measurements.

Hydrous Fo90 olivine single crystals were first observed under a polarising microscope and selected based on the absence of inclusions and their sharp optical extinction. Preliminary X-ray diffraction measurements were carried out on a Huber single-crystal diffractometer equipped with a point detector and MoK $\alpha$  radiation, and driven by the software SINGLE (Angel and Finger, 2011). Sharp diffraction peaks (full width half maxima  $< 0.055^\circ$ ) were observed upon omega-scan rotations for each crystal, confirming that they are of high quality. Two crystals were selected and subsequently oriented parallel to the (101) or (161) crystallographic plane, glued on a glass slide and polished on both sides to obtain platelets (crystals X1 and X2, respectively) with a thickness of  $\sim 15$   $\mu\text{m}$ . The two platelets were then cut into semi-circular or rectangular shapes (Figure 1) using a FEI Scios focused ion beam (Schulze et al., 2017), operated at 30 nA and 30 kV.

High-pressure SCXRD and Brillouin scattering measurements were carried out in a BX-90 piston-cylinder-type diamond anvil cell (DAC) (Kantor et al., 2012) equipped with Almax-Boehler diamond anvils having culets of 500  $\mu\text{m}$  in diameter and conical seats with large opening angles (Boehler and De Hantsetters, 2004). A Re gasket was indented to a thickness of  $\sim 70$   $\mu\text{m}$  and drilled with an infrared laser to obtain the sample chamber. The two FIB-cut crystal platelets were loaded into the same sample chamber together with a ruby sphere (Figure 1) for pressure determination following the

156 calibration reported by Shen et al. (2020). Pre-compressed helium was loaded as quasi-hydrostatic  
157 pressure transmitting medium using the gas loading apparatus installed at the Bayerisches Geoinstitut,  
158 University of Bayreuth (Kurnosov et al., 2008).

159 High-pressure SCXRD and Brillouin scattering measurements were conducted using the system  
160 installed at the Bayerisches Geoinstitut, University of Bayreuth (Trots et al., 2011, 2013). The  
161 Brillouin scattering system consists of a Coherent Verdi V2 solid-state Nd:YVO<sub>4</sub> laser with a 532 nm  
162 single wavelength output and a six-pass Sandercock-type tandem Fabry-Perot interferometer  
163 (Sandercock, 1982) equipped with a Hamamatsu C11202-50 detector. A source laser power of 100  
164 or 150 mW was used for room pressure and high-pressure measurements, respectively. All  
165 measurements were performed in platelet geometry with an external scattering angle of 80°, which  
166 was periodically calibrated using a fused silica glass standard. Dispersion curves of sound wave  
167 velocities versus crystallographic orientation were collected by rotation of the  $\chi$  circle of the  
168 diffractometer between -170° and 180° at steps of 10° or 20°. Densities were derived at each pressure  
169 from single-crystal diffraction measurements, which were performed on the same goniometer using  
170 a Huber Eulerian single-crystal X-ray diffractometer equipped with a point detector. The system is  
171 coupled with an ultra-high intensity rotating anode X-ray source (MoK $\alpha$ , FR-E<sup>+</sup> SuperBright from  
172 Rigaku) operated at 55 kV and 45 mA and multilayer VaryMax<sup>TM</sup> focusing optics, and was driven by  
173 the software SINGLE (Angel and Finger, 2011). At each pressure point, 14-18 Bragg reflections for  
174 each crystal were centred using the eight position centring method (King and Finger, 1979), and cell  
175 parameters were determined by vector least-square refinement (Ralph and Finger, 1982).

## 176 **3. Results and discussion**

### 177 **3.1 Compressibility of hydrous Fo90 olivine**

178 High-pressure SCXRD measurements were conducted at seven pressure points, ranging from  
179 ambient pressure to ~12 GPa. Unit-cell lattice parameters and volumes of both hydrous Fo90 olivine  
180 single crystals investigated in this study are reported in Table 1. The two crystals have slightly  
181 different volumes,  $V(X1) > V(X2)$ , with crystal X1 showing larger uncertainties on the measured



182 lattice parameters. This is due to its crystallographic orientation, which limits the observations in both  
 183  $a^*$  and  $c^*$ . Therefore, lattice parameters measured for crystal X2 are most probably more accurate.  
 184 The variation with pressure of the lattice parameters and volumes, normalized with respect to their  
 185 room pressure values, are reported in Figure 2 and compared with literature data. Hydrous olivine is  
 186 most compressible along the **b**-direction ([010]) and least compressible along the **a**-direction ([100]),  
 187 with the axial compressibility scheme being  $\beta_b > \beta_c > \beta_a$ . The same axial compressibility scheme has  
 188 been also observed for end-member Fo100 (e.g., Downs et al., 1996; Pamato et al., 2019). A third-  
 189 order Birch-Murnaghan Equation of State (BM3 EoS) has been used to fit the  $P$ - $V$  data of both crystals  
 190 using the EoSFit7c program (Angel et al., 2014). The resulting EoS parameters are:  $V_0 = 291.65(2)$   
 191  $\text{\AA}^3$ ,  $K_{T0} = 124.5(1.0)$  GPa and  $K' = 4.5(2)$ . The fitted  $V_0$  value is in excellent agreement (within  $1\sigma$ )  
 192 with the unit-cell volume measured at room pressure for crystal X2. Linearized BM3 EoSs have been  
 193 used to fit the variation with pressure of the  $a$ -,  $b$ - and  $c$ -axis and the resulting EoS parameters are:  $a_0$   
 194  $= 4.7610(1)$   $\text{\AA}$ ,  $M_{0a} = 539.1(5.7)$  GPa,  $M_a' = 23.7(1.4)$ ,  $b_0 = 10.2238(3)$   $\text{\AA}$ ,  $M_{0b} = 283.4(2.8)$  GPa,  $M_b' =$   
 195  $10.7(6)$  and  $c_0 = 5.9921(1)$   $\text{\AA}$ ,  $M_{0c} = 372.1(2.6)$  GPa,  $M_c' = 12.1(5)$ . The axial modulus for the  $a$ -axis  
 196 is much larger than that obtained for the  $b$ -axis, similarly to what is observed for the corresponding  
 197 anhydrous phase Fo90 olivine, as discussed later.

198 Angel et al. (2018) recently reviewed all the published single-crystal data that constrain the elastic  
 199 properties and EoS of mantle-composition olivine (Fo90 to Fo92), also testing the mutual consistency  
 200 of the different datasets used. Therefore, data selected by Angel et al. (2018) will be used as a  
 201 comparison for hydrous olivine. The unit-cell lattice parameters reported by Nestola et al. (2011) and  
 202 Zha et al. (1998), normalized to their corresponding room pressure values, are compared to the single  
 203 crystal hydrous Fo90 olivine data obtained in this study (Figure 2). When compared to the  $P$ - $V$  and  
 204 linearized BM3 EoS fits of the hydrous Fo90 olivine data obtained in this study, the relative  
 205 compression curves (lattice parameters and volumes) for the corresponding anhydrous phase show  
 206 good agreement. Hence, we do not observe anomalies in the axial compressibility scheme or the unit-  
 207 cell volume compression of the hydrous phase.

208

### 209 **3.2 Sound wave velocities of hydrous Fo90 olivine and $c_{ij}$ inversion strategy**

210 Sound wave velocities of hydrous Fo90 olivine were measured at seven pressure points, ranging  
 211 from ambient pressure to ~12 GPa, i.e., the same conditions of SCXRD measurements (Table 2).  
 212 Typical Brillouin spectra collected in the low- and high-pressure range (Figure 3a) show distinct and  
 213 resolved compressional  $v_P$  and shear ( $v_{S1}$  and  $v_{S2}$ ) wave velocities peaks for most ranges of rotation  
 214 angle ( $\chi$ ) (Figure 3b). To obtain the nine elastic stiffness coefficients  $c_{ij}$ , the Christoffel's equation  
 215 (Equation 1) is usually solved using a non-linear least-squares inversion of the density and all the  
 216 acoustic velocities, collected with varying azimuthal angles, of the crystal platelets at each individual  
 217 pressure point. This procedure is referred to as individual fit. Individual fits were performed at each  
 218 pressure point by inverting all the measured acoustic velocities and density data, which were  
 219 calculated self-consistently from the measured unit-cell volumes. Individual fits converged with small  
 220 residuals at each pressure point (Figure 3b) thanks to compressional and shear wave velocities being  
 221 observed for most ranges of rotation angle ( $\chi$ ) and the low correlation between individual elastic  
 222 stiffness coefficients resulting from the choice of the orientations of the two crystal platelets X1 and  
 223 X2 used. Uncertainties in the determined  $c_{ij}$  values are generally less than 1%, except for  $c_{12}$  (< 2%)  
 224 (Table 2).

225 Recently, a non-conventional fitting procedure for Brillouin data (referred to as global fit) was  
 226 proposed by Kurnosov et al. (2017) and described in detail in Buchen (2018). It consists of a global  
 227 inversion of all sound velocity and density data, instead of independently inverting the data measured  
 228 at each individual pressure point. This is practically done by fitting all sound velocity and density  
 229 data with third-order finite strain EoSs describing the evolution with pressure of each  $c_{ij}$ , using the  
 230 formalism of Stixrude and Lithgow-Bertelloni (2005):

$$231 \quad c_{ijkl} = (1 + 2f)^{\frac{5}{2}} \{ c_{ijkl,0} + (3K_0 c'_{ijkl,0} - 5c_{ijkl,0})f + \left( 6K_0 c'_{ijkl,0} - 14c_{ijkl,0} - \frac{3}{2}K_0 \delta_{kl}^{ij} (3K'_0 - 16) \right) f^2 \} \quad (2)$$

232 where  $c_{ijkl}$  is the elastic stiffness coefficient in tensorial notation at a given density (i.e., pressure),  $f$   
 233 is the finite Eulerian strain defined as  $\frac{1}{2} \left[ \left( \frac{\rho_0}{\rho} \right)^{\frac{2}{3}} - 1 \right]$ ,  $c_{ijkl,0}$  is the elastic stiffness coefficient at ambient  
 234 conditions and  $c'_{ijkl,0}$  its pressure derivative,  $K_0$  is the bulk modulus at ambient conditions and  $K'_0$  its  
 235 pressure derivative, and  $\delta^{ijkl}$  is equal to  $-3$  for  $c_{1111}$ ,  $c_{2222}$ , and  $c_{3333}$  and to  $-1$  for the other six  
 236 independent components of the elastic tensor. Thus, by refining the ambient-pressure stiffness  
 237 coefficients ( $c_{ij,0}$ ) and their pressure derivatives ( $c'_{ij,0}$ ), a fit of all measured velocities at all pressure  
 238 points is obtained. This approach has the advantage that all velocity data from all pressure points are  
 239 used to constrain the  $c_{ij}$ s, minimizing the effect of data scattering on the calculated  $c_{ij}$ s and thus  
 240 reducing their estimated uncertainties (Buchen, 2018). Even though this fitting procedure is  
 241 particularly suited for very high pressure data, where some of the  $c_{ij}$ s are poorly constrained due to  
 242 the lack of observations (e.g., Criniti et al., 2021), it can be applied to any high-pressure sound  
 243 velocity dataset and the two procedures should yield consistent results as long as the collected data  
 244 are of high quality, the high-pressure evolution of all  $c_{ij}$  is well described by a third-order finite strain  
 245 EoS, and no phase transition takes place in the investigated pressure interval. Therefore, we also  
 246 applied the global fit procedure to our dataset, which yielded very consistent results compared to  
 247 those obtained by individual fits. The  $c_{ij}$ s calculated from two procedures show virtually no  
 248 discrepancies, with values typically identical within two standard deviations (Table 2, Figure 4).

249 Because density is measured at each pressure point, it is possible to calculate pressure without  
 250 relying on a secondary scale (e.g., ruby) using the relative change in volume of the sample, obtained  
 251 by X-ray diffraction, and the 3<sup>rd</sup>-order finite strain equations described above. From these, an  
 252 expression for the isothermal bulk modulus in the Reuss bound ( $K_{TR}$ ) as a function of volume is  
 253 obtained and the absolute pressure ( $P_{abs}$ ) can be calculated by integrating  $K_{TR}/V$  over a given volume  
 254 interval:

$$255 \quad P_{abs} = \int_{V_0}^V \frac{K_{TR}(V)}{V} dV = 3K_{TR0}f(1+2f)^{\frac{5}{2}} \left[ 1 + \frac{3}{2}(K'_{TR0} - 4)f \right] \quad (3)$$

256 where  $K_{\text{TR}0}$  is the bulk modulus at ambient conditions,  $K'_{\text{TR}0}$  is its pressure derivative,  $f$  is the finite  
 257 Eulerian strain defined as  $\frac{1}{2} \left[ \left( \frac{V}{V_0} \right)^{\frac{2}{3}} - 1 \right]$ . The experimentally determined adiabatic bulk modulus  $K_{\text{SR}}$   
 258 (Table 2) was converted into  $K_{\text{TR}}$  using the relations  $K_{\text{SR}} = K_{\text{TR}}(1 + \alpha\gamma T)$  and  $\alpha = \gamma C_V / K_{\text{TR}} V$ . The  
 259 thermoelastic parameters used for the conversion are  $\theta_D = 644$  K,  $\gamma_0 = 1.044$  and  $q = 1.88$ ,  
 260 corresponding to Fo90 (Angel et al., 2018), and are here assumed to be H<sub>2</sub>O-independent. Then,  $K_{\text{TR}0}$   
 261 and  $K'_{\text{TR}0}$  were calculated by fitting a BM3 EoS to our  $K_{\text{T}}-V$  dataset. A comparison between the  
 262 absolute pressure and the pressures determined from the ruby fluorescence shift is plotted in  
 263 Supplementary Fig. S1, along with the BM3 EoS fits to the  $K_{\text{SR}}-\rho$  datasets obtained from the global  
 264 and individual fits, which show excellent agreement.

265 Figure 4 shows individual  $c_{ij}$  as a function of absolute pressure, as well as selected literature data  
 266 for anhydrous Fo90 olivines for comparison, following the re-analysis of all the published single  
 267 crystal data proposed by Angel et al. (2018) (Supplementary Table S2). The longitudinal moduli ( $c_{11}$ ,  
 268  $c_{22}$  and  $c_{33}$ ), the off-diagonal moduli ( $c_{12}$ ,  $c_{13}$  and  $c_{33}$ ) and the shear moduli  $c_{44}$  and  $c_{66}$  follow a nearly  
 269 linear increase with pressure, whereas the shear modulus  $c_{55}$  exhibits a slightly downward trend  
 270 towards higher pressures. The longitudinal moduli  $c_{11}$ ,  $c_{22}$ ,  $c_{33}$  and the shear moduli  $c_{44}$ ,  $c_{55}$ ,  $c_{66}$  follow  
 271 similar trends with pressure compared to the anhydrous phase; only  $c_{55}$  and  $c_{66}$  are slightly offset to  
 272 lower values in the range of 5 to 10 GPa (Figure 4e). The off-diagonal moduli  $c_{12}$ ,  $c_{13}$ ,  $c_{23}$  are  
 273 characterized by very similar values along the whole pressure interval, with the stiffness scheme being  
 274  $c_{23} > c_{12} > c_{13}$ . Due to the small differences in the magnitude of the off-diagonal moduli, their values  
 275 are frequently the same within uncertainties, especially at high pressure. The off-diagonal moduli  $c_{12}$ ,  
 276  $c_{13}$ ,  $c_{23}$  of the anhydrous phase are scattered but show comparable trends with those of the hydrous  
 277 phase, especially  $c_{23}$ . Even if  $c_{13}$  is generally stiffer than  $c_{12}$ , in contrast with the hydrous phase, their  
 278 systematics is more complex, as either they frequently cross over or are the same within uncertainties.

### 279 280 **3.3 Elastic Properties of Hydrous Fo90 vs Anhydrous Fo90 Olivine**

281 The adiabatic  $K_S$  and  $G$  were calculated from the  $c_{ij}$  values in the Reuss and Voigt bounds and in  
 282 the Voigt-Reuss-Hill average at each experimental pressure point and are reported in Table 2. The  
 283 elastic moduli obtained in this study were then fitted with third-order finite strain EoS to obtain the  
 284 elastic moduli at ambient conditions ( $K_{S0}$  and  $G_0$ ) and their pressure derivatives ( $K'_{S0}$  and  $G'_0$ ) using  
 285 the finite strain EoS of Stixrude and Lithgow-Bertelloni (2005):

$$286 \quad K = (1 + 2f)^{\frac{5}{2}} \left[ K_0 + (3K_0K'_0 - 5K_0)f + \frac{27}{2} (K_0K'_0 - 4K_0)f^2 \right] \quad (4)$$

$$287 \quad G = (1 + 2f)^{\frac{5}{2}} \left[ G_0 + (3K_0G'_0 - 5G_0)f + \left( 6K_0G'_0 - 24K_0 - 14G_0 + \frac{9}{2}K_0K'_0 \right) f^2 \right] \quad (5)$$

288 The EoS parameters for  $K$  and  $G$  in the Voigt and Reuss bound are reported in Table 4. Both  $K_S$  and  
 289  $G$  show a monotonic increase with pressure (Figure 5a) and are perfectly described by third-order  
 290 EoS.

291 To quantify the effect of 0.20 wt.%  $H_2O$  on the Fo90 elastic properties, the EoS parameters for  
 292 hydrous Fo90 olivine determined in this study have to be compared to those of the corresponding  
 293 anhydrous phase. The elastic behaviour of anhydrous Fo90 olivine has been largely investigated in  
 294 the past decades and its full elastic tensor has been constrained by a variety of techniques. Angel et  
 295 al. (2018) presented a re-analysis of all available single crystal data for mantle-composition olivine  
 296 (Fo90-92) providing best-fit EoS parameters, which were recently used to calculate olivine density  
 297 profiles over the upper mantle under different thermal regimes (Faccincani et al., 2021). However, a  
 298 comparison of the EoS parameters determined in this study with those from Angel et al. (2018) cannot  
 299 be made for two main reasons: (i) a best fit of the shear modulus data is missing, (ii) the fit includes  
 300 high-temperature volume data and high-pressure, high-temperature (HP-HT) elasticity data, which  
 301 are not available for hydrous Fo90 olivine. For this reason, we refitted the high-pressure elasticity  
 302 data ( $K_S$  and  $G$ ) for anhydrous Fo90 originally used in the fit from Angel et al. (2018) with the same  
 303 formalism used for hydrous Fo90 olivine (Equations 4 and 5). The data used correspond to Fo90  
 304 olivines, and were selected up to ~14 GPa, i.e., the uppermost limit of olivine stability field. In our  
 305 examination, we also made a re-analysis of the available shear modulus data to provide the best fit  
 306 for  $G_0$  and  $G'_0$  of anhydrous Fo90 olivine.  $K_S$  and  $G$  were recalculated from the  $c_{ij}$  values reported at

307 high pressure by Abramson et al. (1997) where the data are complete (i.e., four pressure points), the  
308 data of Zha et al. (1998) (i.e., four pressure points), the data of Mao et al. (2015) (i.e., seven pressure  
309 points) and the data of Zhang and Bass (2016) (i.e., five pressure points) (Figure 5b, Supplementary  
310 Table S2). The EoS parameters for  $K_S$  and  $G$  in the Voigt and Reuss bound are reported in Table 4.

311 The EoS parameters for hydrous and anhydrous Fo90 olivine at ambient conditions are marginally  
312 different (Table 4). The incorporation of 0.20 wt.%  $H_2O$  into the olivine crystal structure causes a  
313 reduction in  $K_{S0}$  and  $G_0$  of ~1.2% and ~1.6%, respectively, and an increase in  $K'_{S0}$  and  $G'_0$  of ~2.9%  
314 and ~4.5%, respectively. Therefore, the elastic moduli of hydrous Fo90 olivine at ambient pressure  
315 are relatively softer compared to those of anhydrous Fo90 but are characterized by larger derivatives.  
316 Indeed,  $K$  and  $G$  of hydrous Fo90 increase more rapidly with pressure so that the elastic behaviour of  
317 hydrous and anhydrous Fo90 olivine becomes indistinguishable within uncertainties at pressures  
318 exceeding ~3-4 GPa. This is consistent with a recent high-pressure single-crystal X-ray diffraction  
319 study conducted on a sample of Fo95 olivine with a low water content (~0.15 wt.%  $H_2O$ ) (Xu et al.,  
320 2020), where hydrogen substitution is predominantly associated with the T site. As the effect of water  
321 on the elastic moduli and their derivatives in our Fo90 and Fo100 (Mao et al., 2010) may also depend  
322 on different H substitution mechanisms in olivine, besides distinct water contents, comparisons  
323 among different data should be made carefully. The Fo100 single crystals studied by Mao et al. (2010)  
324 were synthesized by Smyth et al. (2006), run SZ0408A, to which the reader is referred for synthesis  
325 conditions and FTIR spectrum. For this sample, it was suggested that hydration predominantly  
326 occurred through the octahedral substitution  $Mg^{2+} \leftrightarrow 2H^+$ . On the other hand, a tetrahedral  
327 substitution  $Si^{4+} \leftrightarrow 4H^+$  has been suggested for our sample (Withers et al., 2011). Note, however, that  
328 the FTIR spectra of hydrous Fo100 (Smyth et al., 2006, Figure 2) and hydrous Fo90 (Withers et al.,  
329 2011, Figure 2) both show strong absorbance features in the identical high wavenumber region,  
330 hinting that the two samples should present the same substitution mechanism. Various pieces of  
331 evidence suggest that absorption features in the high wavenumber region (~3.450-3.600  $cm^{-1}$ ) are  
332 related to OH bonding to T sites (Fei and Katsura, 2020; Umemoto et al., 2011; Xue et al., 2017),

therefore we may expect that both the Fo100 and Fo90 sample contain predominantly Si vacancies. The important observation, however, for the aim of this study is that both samples present the same type of defects and therefore we can compare the effect of hydration on their elastic properties directly, upon normalization with respect to H<sub>2</sub>O concentration. The effect of water on the elastic parameters can be then expressed as:

$$\Delta_{H_2O}M(\%/wt.\% H_2O) = \frac{M_{hydrus}-M_{dry}}{M_{dry}} \times \frac{1}{X_{H_2O}(wt.\%)} \times 100\% \quad (6)$$

where  $M$  can be  $K_{S0}$ ,  $G_0$  (in GPa),  $K'_{S0}$  or  $G'_{S0}$  (non-dimensional) and  $X_{H_2O}$  is the water content of the hydrous sample in wt.%. The obtained results for both Fo90 and Fo100 are reported in Table 4.  $\Delta_{H_2O}M$  values are affected by large uncertainties, especially in the case of Fo90. This mostly arises from the fitting parameters of anhydrous Fo90, which are less well-constrained because four different datasets were combined in the EoS fit of the elastic moduli (Table 4).  $\Delta_{H_2O}K_{S0}$  and  $\Delta_{H_2O}G_{S0}$  for Fo90 and Fo100 are on the margins of being mutually consistent. As predicted by Jacobsen et al. (2008, 2009),  $K_{S0}$  and  $G_0$  of Fo90 are slightly more reduced by H<sub>2</sub>O compared to Fo100. However,  $\Delta_{H_2O}K'_{S0}$  and  $\Delta_{H_2O}G'_{S0}$  values are in excellent agreement within mutual uncertainties (Table 4). Therefore, the more pronounced effect observed by Mao et al. (2010) on hydrous Fo100 elastic properties arises by virtue of the higher water content, and not from a different effect of water in Fe-free and Fe-bearing samples as shown in Table 4. This also suggests that hydration is most likely to have a linear effect on the pressure derivatives of olivines.

351

### 3.4 Effect of Hydration on Sound Wave Velocities of Fo90 Olivine and its Geophysical Implications

Using the new experimental data obtained in this study, we calculated the aggregate sound wave velocities of hydrous Fo90 olivine with 0.20 wt.% H<sub>2</sub>O as a function of pressure. Aggregate sound wave velocities of anhydrous Fo90 olivine are compared with those for hydrous olivine (Figure 6). As the effect of temperature on the elasticity of hydrous olivine is currently unknown, all the calculations were performed at 300 K. The effect of water on the thermal expansion of hydrous (~2.5

359 wt.% H<sub>2</sub>O) wadsleyite and ringwoodite, i.e., the high-pressure polymorphs of olivine, seems to be  
360 negligible (Inoue et al. 2004). Therefore, in virtue of the comparatively lower water content of our  
361 hydrous olivine, H<sub>2</sub>O is not expected to have a significant effect on the thermal expansion of olivine.

362 The  $v_P$  and  $v_S$  of hydrous Fo90 follow similar trends to those of the elastic moduli (Figure 6), with  
363 velocities being lower than in anhydrous Fo90 by 0.8% and 0.9%, respectively, at ambient conditions  
364 and becoming the same within uncertainty above 5 GPa. Mao et al. (2010) showed that  $v_P$  and  $v_S$  of  
365 the hydrous Fo100 cross and exceed those of the corresponding anhydrous phase at about 4 and 3  
366 GPa, respectively, and rapidly diverge at higher pressure. The prominent rise of  $v_P$  and  $v_S$  with  
367 increasing pressure is due to the higher water content of the hydrous Fo100 sample employed by Mao  
368 et al. (2010) and not to a different effect of water in Fe-free and Fe-bearing samples, as shown in  
369 Table 4.

370 The 410-km global seismic discontinuity is widely accepted to be caused by the phase transition  
371 of olivine to its high pressure polymorph wadsleyite (Frost, 2008). Several works attempted to  
372 constrain the bulk olivine content in the upper mantle by comparing sound wave velocities of olivine  
373 and wadsleyite, calculated from either experimental or computational mineral physics data, with the  
374 observed seismic velocity contrasts across the discontinuity (e.g., Bass and Anderson, 1984;  
375 Dziewonski and Anderson, 1981; Núñez-Valdez et al., 2013; Wang et al., 2014, 2019). However, the  
376 calculated velocity contrasts across the discontinuity for a pyrolite composition (~60 vol.% olivine)  
377 are not consistent with global 1-D seismic models (e.g., Preliminary Earth Reference Model, PREM,  
378 or AK135; Dziewonski and Anderson, 1981; Kennett et al., 1995), pointing towards a bottom upper  
379 mantle that is less olivine-rich than pyrolite. Considering the reduction of the sound wave velocities  
380 of wadsleyite due to the incorporation of water (Mao et al., 2008a, 2008b), it was proposed that water  
381 dissolved in olivine and wadsleyite may reconcile the pyrolite model with seismological observations.  
382 Water is preferentially partitioned in wadsleyite ( $D_{\text{wad/ol}} = 6$ ; Thio et al., 2016) and whether the  
383 presence of water can resolve seismological observations with pyrolitic mantle olivine contents will  
384 depend on both the expected amount of water in olivine and to what extents it affects  $v_P$  and  $v_S$  of



385 olivine. Available experimental data for hydrous olivine indicate storage capacities up to 0.9 wt.%  
386 (Smyth et al., 2006), but the water content of olivine under relevant deep upper mantle conditions  
387 will be much lower, around 0.2 – 0.5 wt.% H<sub>2</sub>O (e.g., Férot and Bolfan-Casanova, 2012; Hirschmann  
388 et al., 2005; Mosenfelder, 2006). In this regard, our data are particularly significant and indicate that  
389 the incorporation of ~0.20 wt.% H<sub>2</sub>O into Fo90 olivine crystal structure does not significantly affect  
390 its  $v_P$  and  $v_S$  at high pressure. To explore whether the presence of higher water contents in olivine  
391 would cause greater effects on its  $v_P$  and  $v_S$ , we used equation (6) to calculate the sound velocities of  
392 Fo90 hosting 0.5 wt.% H<sub>2</sub>O (Table 4, Figures 5c-6c). Because hydration causes an increase in the  
393 unit-cell volume, we recalculated the room pressure volume of Fo90 with 0.5 wt.% H<sub>2</sub>O using the  
394 factor  $+5.5 [A^3] \times 10^{-5} \times \text{H}_2\text{O} [\text{ppm}]$  according to the expression from Smyth et al. (2006), and then  
395 converted the resulting volume into density. The calculated sound wave velocities for Fo90 with 0.5  
396 wt.% H<sub>2</sub>O are reported in Figure 6c and compared to those of hydrous Fo90 with 0.20 wt.% H<sub>2</sub>O and  
397 anhydrous Fo90. Although the incorporation of 0.5 wt.% H<sub>2</sub>O into Fo90 olivine strongly reduces its  
398 elastic moduli and sound wave velocities at ambient pressure (Table 4, Figures 5c-6c),  $v_P$  and  $v_S$  of  
399 the hydrous phase become indistinguishable within uncertainties with those of the anhydrous phase  
400 at pressures exceeding ~9 and ~12 GPa, owing to the larger  $K'_{S0}$  and  $G'_0$ . This advises caution when  
401 speculating the water content in the deep upper mantle based on its effect on olivine elastic properties  
402 and seismic wave velocities. Furthermore, our data also suggest that H<sub>2</sub>O incorporation in olivine  
403 may not reconcile seismological observations at the 410-km discontinuity with a pyrolitic mantle,  
404 although we are aware that direct determinations of sound wave velocities of hydrous olivine and  
405 hydrous wadsleyite at combined HP–HT (e.g., Buchen et al., 2018) are needed to refine these  
406 findings.

407 The discrepancy between the observed and calculated wave velocities for a pyrolite composition  
408 may also arise from strong variations of olivine content and upper mantle lithologies near the 410-  
409 km seismic discontinuity, for which corroborating evidence has been found by Zhang and Bass  
410 (2016). Indeed, olivine contents inferred from regional seismic models of the Pacific region are

411 extremely variable and increase from approximately 20 – 40 % in the central Pacific to 60 – 90 % in  
412 the western U.S. and eastern Pacific regions. Given the indistinguishable seismic behaviour of  
413 hydrous and anhydrous Fo90 olivine at high pressure, we argue that this high degree of heterogeneity  
414 does not stem from the potentially different responses of dry and wet regions of the deep upper mantle,  
415 but rather may arise from actual variations in olivine content.

#### 416 **4. Concluding remarks**

417 The sound velocities and single-crystal elastic coefficients of Fo90 olivine with 0.20 wt.% H<sub>2</sub>O  
418 were measured up to ~12 GPa at room temperature by simultaneous single-crystal X-ray diffraction  
419 and Brillouin scattering experiments. Compared to the anhydrous phase, *K* and *G* of hydrous Fo90 at  
420 ambient conditions are slightly offset to lower values, while their pressure derivatives are slightly  
421 larger. Nonetheless, the elastic behaviour of hydrous and anhydrous Fo90 olivine becomes  
422 indistinguishable within uncertainties at pressures corresponding to the base of the upper mantle.  
423 Using our new accurate data, we investigated the effect of hydration on aggregate sound velocities of  
424 Fo90. At ambient pressure, the compressional and shear wave velocities of hydrous Fo90 with 0.2 –  
425 0.5 wt.% H<sub>2</sub>O are slightly slower compared to those of the hydrous phase, but become  
426 indistinguishable within uncertainties at deep upper mantle conditions. Therefore, if amounts of water  
427 were to be incorporated into Fo90 olivine crystal structure, its elastic and seismic behaviour at high  
428 pressure may remain unchanged. Based on our findings, we suggest that water in olivine is not  
429 seismically detectable, at least for contents up to 0.2 – 0.5 wt.%, i.e., the amount of water expected  
430 in olivine at deep upper mantle conditions. We therefore advise caution about speculations of the  
431 water content in the deep upper mantle based on its effect on olivine elastic properties and sound  
432 wave velocities. In addition, our data also suggest that the hydration of olivine is unlikely to be a key  
433 factor in reconciling seismic velocity and density contrasts across the 410-km discontinuity with a  
434 pyrolitic mantle.

435 **Author contribution statement**

436 [L.F.] Conceptualization, Methodology, Formal analysis, Investigation, Data Curation, Writing–  
437 Original Draft, Writing–Review & Editing; [G.C.]: Methodology, Formal analysis, Investigation,  
438 Data Curation, Writing–Review & Editing; [A.K.]: Methodology, Investigation, Writing–Review &  
439 Editing; [T.B.B.]: Methodology, Investigation, Writing–Review & Editing, Supervision; [A.W.]:  
440 Investigation, Writing–Review & Editing; [M.M.]: Writing–Review & Editing, Supervision; [F.N.]:  
441 Writing–Review & Editing, Supervision; [M.C.]: Conceptualization, Writing–Review & Editing,  
442 Supervision.

443 **Declaration of Competing Interest**

444 The authors declare that they have no known competing financial interests or personal  
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454 **Data Availability**

455 All data derived from this research are presented in the enclosed tables, figures, and  
456 supplementary material.

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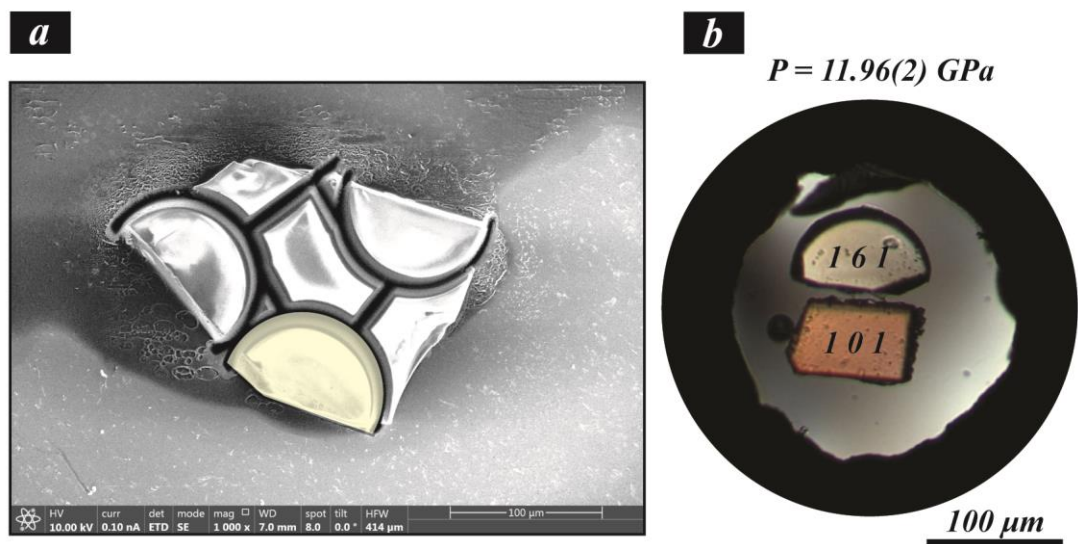
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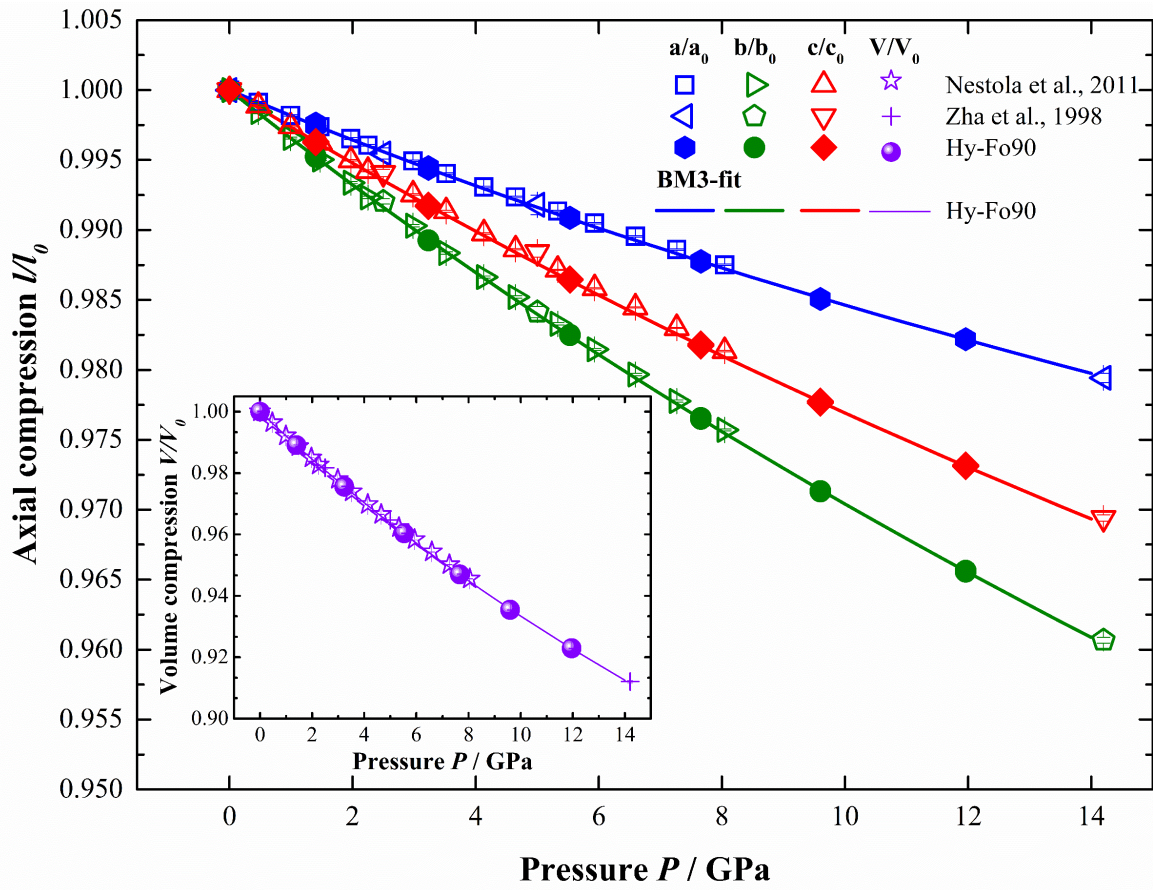
665 **Figures**

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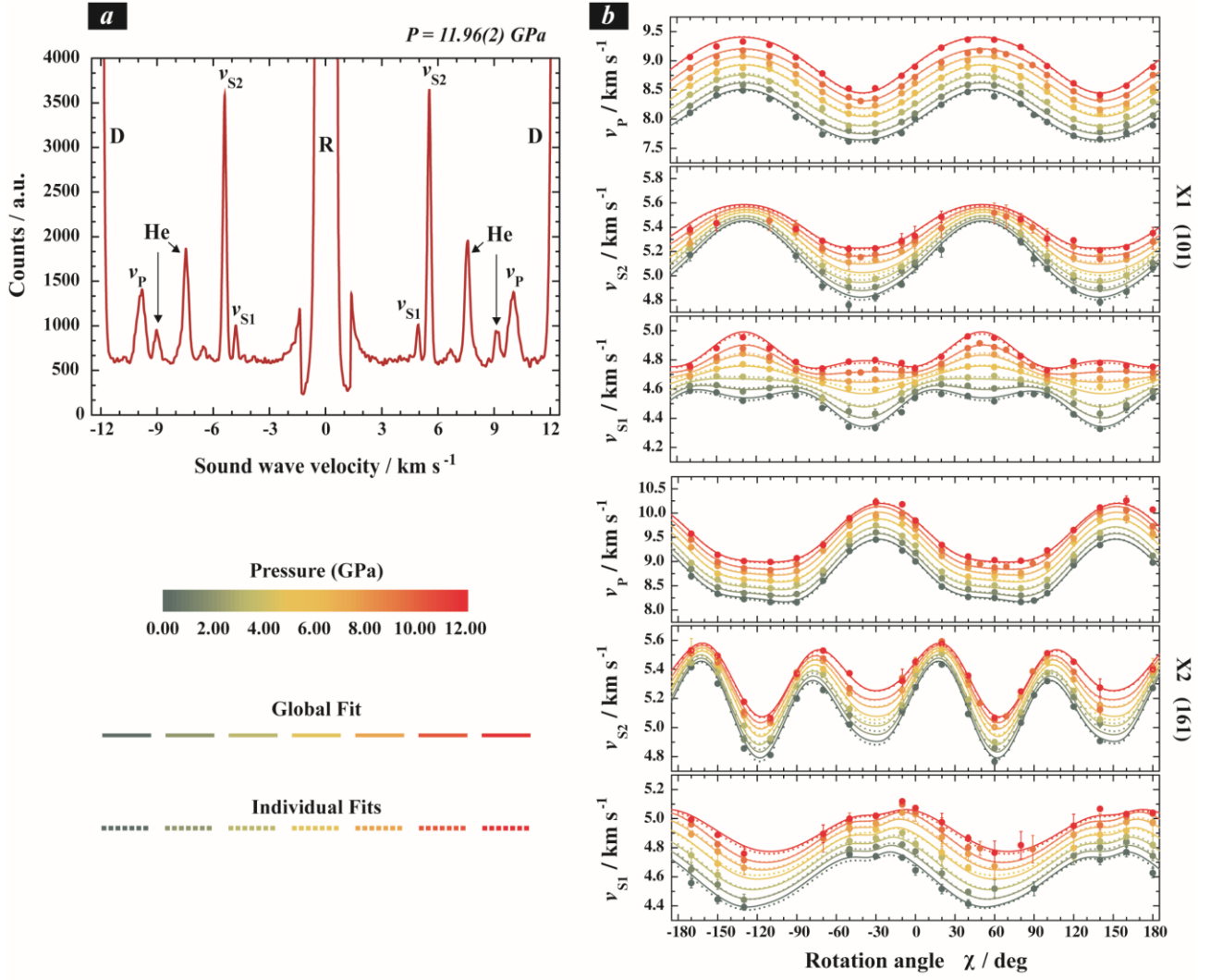
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668 **Figure 1.** (a) Secondary electron image of a Fo90 olivine platelet oriented parallel to the (161) plane  
669 (crystal X2) after the FIB cutting procedure; a superimposed yellow semicircle denotes the sample  
670 that was then loaded in the DAC. (b) Cross-polarized light photomicrographs of platelet X2 (161)  
671 and X1 (101) inside the sample chamber of the DAC at high pressure, together with a ruby chip (dark  
672 black sphere on the left of 101 platelet).



673

674 **Figure 2.** Unit-cell parameters and unit-cell volumes (inset) of crystals X1 and X2 normalized with  
675 respect to their room pressure values; data were fitted with a third-order Birch-Murnaghan EoS using  
676 the EosFit7c program (Angel et al., 2014). Literature data of anhydrous Fo90-92 olivines (Nestola et  
677 al., 2011; Zha et al., 1998; Supplementary Table S1) are shown for reference. Note the good  
678 agreement between the BM3 fits and relative compression data of hydrous Fo90 olivine and the  
679 literature data for the corresponding anhydrous phase.



680

681 **Figure 3.** (a) Selected Brillouin spectrum of crystal X2 (161) at 11.96(2) GPa showing distinct and

682 well-resolved compressional  $v_P$ , slow and fast shear  $v_{S1}$  and  $v_{S2}$  wave velocities peaks, as well as

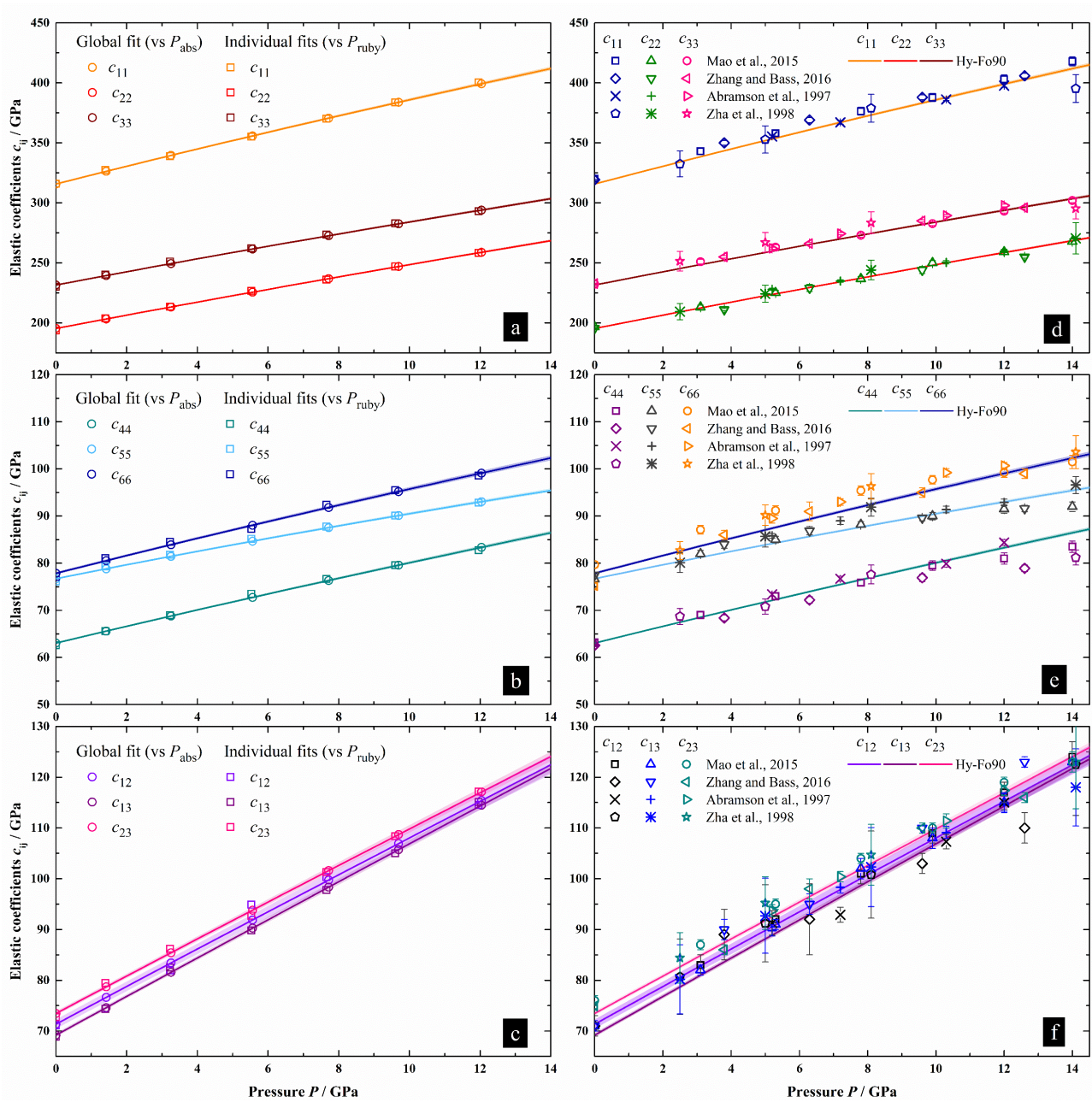
683 spectral contributions of the diamond anvils (D), pressure medium (He) and elastic scattering (R). (b)

684 Data points (filled symbols) as a function of the rotation angle ( $\chi$ ) for both platelets and dispersion

685 curves obtained from the global fit (solid lines) and individual fits (dashed lines), showing excellent

686 agreement between the two fitting strategies.





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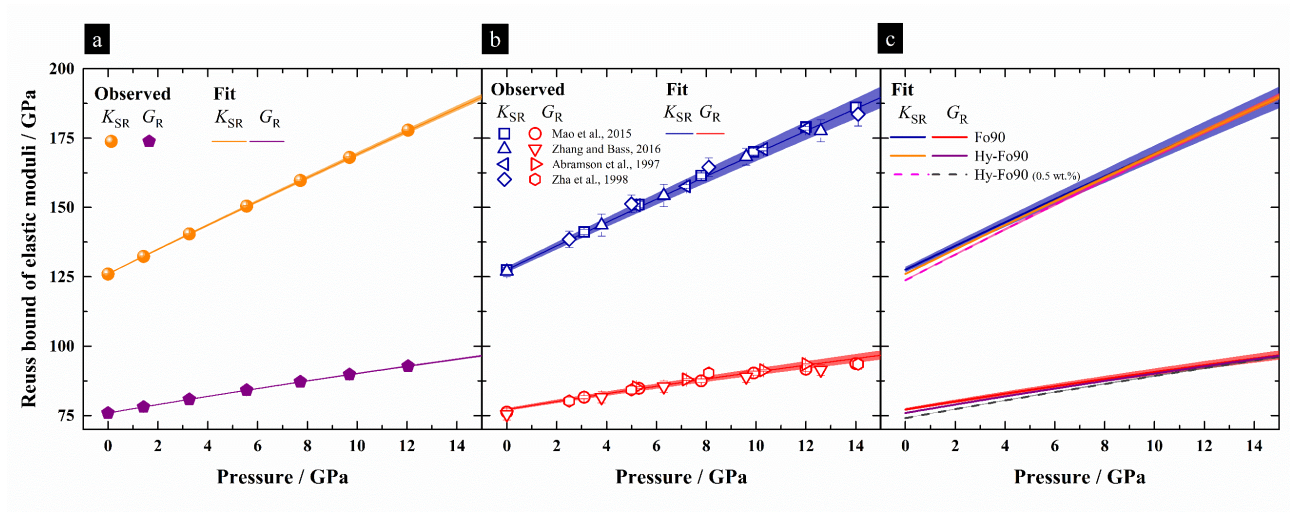
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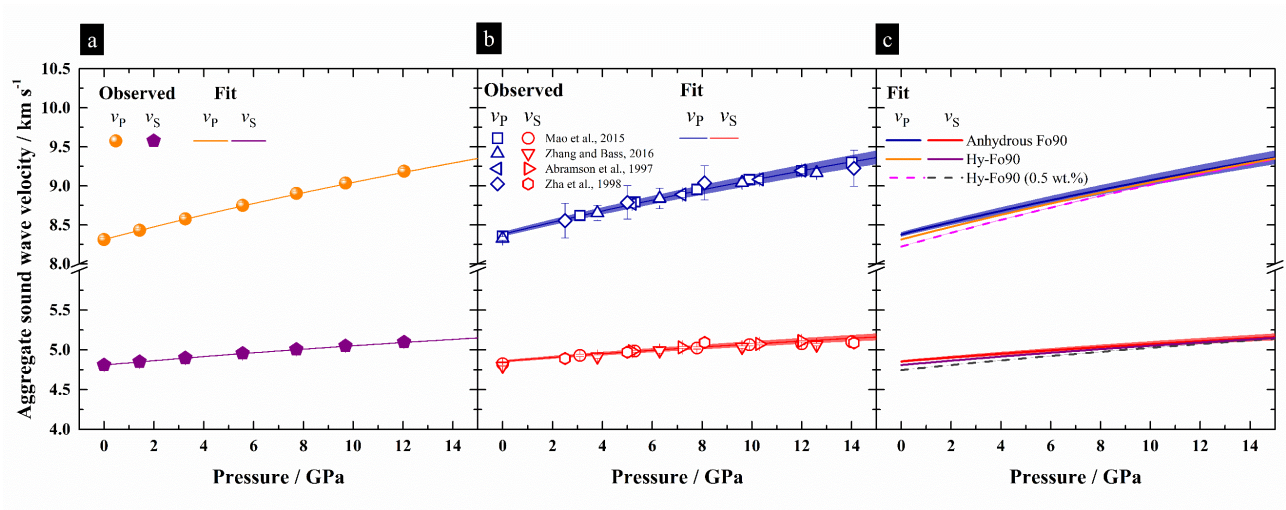
**Figure 4.** (a-b-c) Comparison of the  $c_{ij}$  obtained through the global fit (open circles) and individual fits (open squares), showing values typically identical within two standard deviations (Table 2); the solid lines represent 3rd order EoS fits of each  $c_{ij}$  (global fit parameters) (Table 3). (d-e-f) Comparison of the  $c_{ij}$  of anhydrous Fo90 olivines selected from previous literature (Abramson et al., 1997; Mao et al., 2015; Zha et al., 1998; Zhang and Bass, 2016; Supplementary Table S2), and the fits of each  $c_{ij}$  (global fit parameters) of hydrous Fo90 olivine obtained in this study. Note that the fits of the experimentally determined  $c_{ij}$ s for hydrous olivine and the  $c_{ij}$ s of the corresponding anhydrous phase are nearly identical.



696

697 **Figure 5.** (a) Variation as a function of pressure of the bulk ( $K$ ) and shear ( $G$ ) moduli of hydrous  
698 Fo90 calculated in the Reuss bound from the experimental high-pressure elasticity measurements of  
699 this study; the solid line and shaded area are the fit of the experimental data and the associated  
700 uncertainty, respectively. (b) Variation as a function of pressure of the bulk ( $K$ ) and shear ( $G$ ) moduli  
701 of anhydrous Fo90 calculated in the Reuss bound from previous experimental high-pressure elasticity  
702 measurements (Abramson et al., 1997; Mao et al., 2015; Zha et al., 1998; Zhang and Bass, 2016;  
703 Supplementary Table S2); the solid line and shaded area are the fit of the experimental data and the  
704 associated uncertainty, respectively. (c) Superimposition of fit curves for hydrous Fo90 and  
705 anhydrous Fo90 olivine; the dashed lines represent a linear extrapolation of the effect of incorporation  
706 of 0.5 wt. % H<sub>2</sub>O on the elastic properties of Fo90 olivine (see text and Table 4). Uncertainties are  
707 calculated by propagating the experimental errors on density and elastic moduli.





**Figure 6.** (a) Variation as a function of pressure of the aggregate compressional and shear wave velocities of hydrous Fo90 calculated from the experimental high-pressure elasticity measurements of this study; the solid line and shaded area the fit of the experimental data and the associated uncertainty, respectively. (b) Variation as a function of pressure of the aggregate compressional and shear wave velocities of anhydrous Fo90 calculated from previous experimental high-pressure elasticity measurements (Abramson et al., 1997; Mao et al., 2015; Zha et al., 1998; Zhang and Bass, 2016; Supplementary Table S2); the solid line and shaded area the fit of the experimental data and the associated uncertainty, respectively. (c) Superimposition of fit curves for hydrous Fo90 and anhydrous Fo90 olivine. Uncertainties are calculated by propagating the experimental errors on density and elastic moduli.

719 **Tables**

720

721 **Table 1.** Unit-cell lattice parameters and volumes of hydrous Fo90 olivine crystals X1 and X2 measured in the DAC experiment. Pressure is calculated  
722 using the ruby fluorescence calibration of Shen et al. (2020).

X1 (platelet 1 0 1)					X2 (platelet 1 6 1)			
P (GPa)	a (Å)	b (Å)	c (Å)	V (Å <sup>3</sup> )	a (Å)	b (Å)	c (Å)	V (Å <sup>3</sup> )
0.00010(1)	4.7613(5)	10.2243(2)	5.9925(3)	291.72(3)	4.7610(1)	10.2227(3)	5.9921(1)	291.63(1)
1.40(2)	4.7499(7)	10.1753(3)	5.9701(5)	288.55(4)	4.7487(3)	10.1744(5)	5.9700(3)	288.44(2)
3.23(2)	4.7351(5)	10.1147(3)	5.9432(4)	284.64(3)	4.7342(2)	10.1128(4)	5.9423(2)	284.49(2)
5.53(4)	4.7178(7)	10.0455(3)	5.9116(5)	280.16(3)	4.7176(2)	10.0436(4)	5.9108(2)	280.06(2)
7.66(2)	4.7029(6)	9.9847(3)	5.8837(4)	276.28(3)	4.7028(3)	9.9824(7)	5.8826(3)	276.16(3)
9.60(4)	4.6902(7)	9.9313(3)	5.8592(6)	272.92(3)	4.6899(3)	9.9292(5)	5.8581(2)	272.80(2)
11.96(2)	4.6765(6)	9.8731(2)	5.8319(5)	269.27(3)	4.6762(2)	9.8707(4)	5.8309(1)	269.14(1)

723

724 **Table 2.** Elastic stiffness coefficients ( $c_{ij}$ ) and elastic moduli obtained from the individual fit and global fit procedure, reported in GPa. Numbers in  
 725 brackets represent one standard deviation (std).  $\rho$  uncertainties propagate both the std's of the unit-cell volumes from SCXRD and the chemical  
 726 composition from EPMA.

**Individual Fits**

P (GPa)	$\rho$ (g cm <sup>-3</sup> )	$c_{11}$	$c_{22}$	$c_{33}$	$c_{44}$	$c_{55}$	$c_{66}$	$c_{12}$	$c_{13}$	$c_{23}$	$K_{SV}$	$G_V$	$K_{SR}$	$G_R$	$K_{S\ VHR}$	$G_{VHR}$
0.00010(1)	3.345(6)	316.1(9)	193.7(6)	230.2(6)	62.6(2)	76.1(2)	77.1(3)	70.8(1.1)	68.8(5)	72.8(5)	129.4(4)	78.3(1)	125.2(4)	75.4(1)	127.3(4)	76.9(1)
1.40(2)	3.382(6)	327.3(6)	203.6(3)	240.2(5)	65.6(2)	79.3(2)	81.1(2)	74.3(1.0)	74.4(6)	79.5(4)	136.4(4)	81.4(1)	132.3(4)	78.4(1)	134.3(4)	79.9(1)
3.23(2)	3.428(6)	339.0(6)	213.4(4)	251.1(5)	69.0(3)	81.8(2)	84.5(3)	82.0(9)	82.5(4)	86.2(4)	145.0(3)	83.9(1)	140.7(3)	81.1(1)	142.8(3)	82.5(1)
5.53(4)	3.483(6)	355.1(1.1)	226.7(4)	262.1(6)	73.5(2)	85.2(5)	87.2(5)	94.9(1.8)	89.8(7)	92.7(7)	155.4(5)	87.0(1)	150.9(5)	84.4(1)	153.2(5)	85.7(1)
7.66(2)	3.532(6)	370.3(1.0)	236.0(4)	273.4(8)	76.7(2)	87.8(2)	92.4(3)	100.3(1.3)	97.8(6)	101.3(6)	164.3(4)	90.0(1)	159.6(4)	87.4(1)	162.9(4)	89.2(1)
9.60(4)	3.575(6)	383.9(1.0)	247.0(5)	282.9(7)	79.6(2)	90.1(3)	95.5(3)	108.3(1.5)	105.1(6)	108.4(7)	173.0(5)	92.5(1)	168.2(4)	89.8(1)	170.6(4)	91.2(1)
11.96(2)	3.624(6)	400.4(1.4)	258.4(8)	293.0(1.0)	82.8(3)	92.8(3)	98.6(4)	114.5(1.6)	115.2(8)	117.2(8)	182.8(5)	95.2(1)	177.7(5)	92.4(1)	180.3(5)	93.8(1)

**Global Fit**

0.00010(1)	3.345(6)	315.7(7)	193.3(4)	231.5(5)	63.0(2)	76.7(2)	77.8(3)	71.3(9)	69.2(4)	73.5(4)	130.1(3)	78.8(1)	126.0(2)	75.9(1)	128.0(3)	77.3(1)
1.424(7)	3.382(6)	326.3(7)	203.2(4)	239.4(5)	65.6(2)	78.8(2)	80.5(3)	76.6(9)	74.6(4)	78.7(4)	136.5(3)	80.9(1)	132.4(2)	78.1(1)	134.4(3)	79.5(1)
3.26(2)	3.428(6)	339.6(8)	213.2(4)	249.4(5)	68.8(2)	81.5(2)	83.9(3)	83.4(1.0)	81.6(4)	85.4(5)	144.8(3)	83.6(1)	140.4(3)	80.8(1)	142.6(3)	82.2(1)
5.56(3)	3.483(6)	355.8(9)	225.6(5)	261.5(6)	72.7(2)	84.7(2)	88.1(3)	91.9(1.1)	90.3(5)	93.8(5)	155.0(4)	86.9(1)	150.5(3)	84.2(1)	152.7(3)	85.5(1)
7.72(4)	3.532(6)	370.6(1.0)	236.8(6)	272.6(7)	76.3(3)	87.6(3)	91.8(4)	99.7(1.3)	98.3(6)	101.6(6)	164.4(4)	89.8(1)	159.7(3)	87.1(1)	162.0(4)	88.5(2)
9.69(5)	3.575(6)	383.9(1.2)	246.9(7)	282.5(9)	79.6(3)	90.1(3)	95.2(4)	106.9(1.5)	105.7(7)	108.7(7)	172.9(5)	92.4(1)	168.1(3)	89.8(1)	170.5(4)	91.1(2)
12.04(7)	3.624(6)	399.3(1.4)	258.8(8)	294.0(1.0)	83.4(3)	93.1(3)	99.1(5)	115.4(1.7)	114.5(8)	117.1(8)	182.9(5)	95.4(1)	177.9(4)	92.8(1)	180.4(5)	94.1(2)

728     **Table 3.** Resulting fit parameters of the third–order finite strain expression for the  $c_{ij}$  and  $c_{ij}'$  shown in Figure 4.

	$c_{ij}$	$c_{ij}'$
$c_{11}$	315.7(7)	7.21(9)
$c_{22}$	195.3(4)	5.41(5)
$c_{33}$	231.5(5)	5.41(6)
$c_{44}$	63.0(2)	1.74(2)
$c_{55}$	76.7(2)	1.46(2)
$c_{66}$	77.8(3)	1.84(3)
$c_{12}$	71.3(9)	3.59(11)
$c_{13}$	69.2(4)	3.68(5)
$c_{23}$	73.5(4)	3.56(5)

729

730 **Table 4.** EoS parameters for  $K$  and  $G$  in the Voigt and Reuss bound for hydrous and anhydrous Fo90 olivine.

<i>Anhydrous Fo90 olivine – best fit</i>								
	$K_{SV}$		$G_V$		$K_{SR}$		$G_R$	
$M_0$ (GPa)	131.8(1.0)		80.2(5)		127.5(1.0)		77.2(5)	
$M'_0$	4.29(14)		1.42(5)		4.33(14)		1.48(6)	
<i>This study – Hydrous Fo90 olivine (global fit parameters)</i>								
	$K_{SV}$	<i>Effect of H<sub>2</sub>O (%)</i>	$G_V$	<i>Effect of H<sub>2</sub>O (%)</i>	$K_{SR}$	<i>Effect of H<sub>2</sub>O (%)</i>	$G_R$	<i>Effect of H<sub>2</sub>O (%)</i>
$M_0$ (GPa)	130.1(2)	–1.3	78.76(8)	–1.8	126.0(2)	–1.2	75.92(8)	–1.7
$M'_0$	4.41(4)	2.8	1.49(1)	4.9	4.46(4)	3.0	1.54(1)	4.1
<i>Hydrous Fo90 olivine – extrapolation to 0.5 wt.% H<sub>2</sub>O</i>								
	$K_{SV}$	<i>Effect of H<sub>2</sub>O (%)</i>	$G_V$	<i>Effect of H<sub>2</sub>O (%)</i>	$K_{SR}$	<i>Effect of H<sub>2</sub>O (%)</i>	$G_R$	<i>Effect of H<sub>2</sub>O (%)</i>
$M_0$ (GPa)	127.6(2)	–3.2	76.60(8)	–4.5	123.8(2)	–2.9	74.00(8)	–4.1
$M'_0$	4.59(4)	7.0	1.60(1)	12.3	4.66(4)	7.5	1.63(1)	10.1
$\Delta_{H_2O}M$ (%/wt.% H <sub>2</sub> O) – Hydrous Fo90 olivine								
	$K_{SV}$		$G_V$		$K_{SR}$		$G_R$	
$M_0$ (GPa)	–6.4(4.0)		–9.0(3.4)		–5.9(4.1)		–8.3(3.5)	
$M'_0$	14.0(17.5)		24.7(19.2)		15.0(17.4)		20.3(21.6)	
$\Delta_{H_2O}M$ (%/wt.% H <sub>2</sub> O) – Hydrous Fo100 olivine [data from Mao et al., 2010 and Zha et al., 1996]								
	$K_{SV}$		$G_V$		$K_{SR}$		$G_R$	
$M_0$ (GPa)	–3.1(0.9)		–3.2(0.7)		–2.2(0.9)		–3.2(0.7)	
$M'_0$ <sup>1</sup>	7.9(5.9)		27.8(11.1)		7.9(5.9)		27.8(11.1)	

731 <sup>1</sup> We assumed that  $M'_0$  in the Voigt-Reuss-Hill bound  $\approx M'_0$  in the Voigt and Reuss bounds