Single-crystal elastic properties of Ca$_{0.07}$Mg$_{1.93}$Si$_2$O$_6$ orthopyroxene

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ABSTRACT

The single-crystal elastic properties of Ca$_{0.07}$Mg$_{1.93}$Si$_2$O$_6$ orthopyroxene (space group Pbca) have been investigated by Brillouin spectroscopy at ambient conditions. The aggregate bulk and shear moduli, $K_{0.5} = 102.5$ GPa (1.5) and $\mu = 74.2$ GPa (1.1), respectively, are ~5% and ~3% lower than commonly accepted values for MgSiO$_3$ end-member ($K_{0.5} = 107.6$, $\mu = 76.8$ GPa). These results indicate that the incorporation of small amount of Ca in the orthoenstatite structure does not greatly affect its elastic properties. As a consequence, the increase in bulk modulus reported in natural orthopyroxenes relative to the Mg-end-member is not related to the substitution of Ca in the M2 octahedral sites, but more probably to the substitution of Al in tetrahedral sites.

Keywords: Brillouin spectroscopy, elastic properties, enstatite, orthopyroxene

INTRODUCTION

Orthopyroxene (Opx) is a major component in the crust and in mineralogical models of the Earth’s upper mantle to a depth of about 350 km (e.g., Ringwood 1975; Bass and Anderson 1984), where it transforms to a high-pressure monoclinic polymorph (e.g., Pacalo and Gasparik 1990; Kanzaki 1991). Knowledge of its elastic properties is therefore important to construct reliable compositional and mineralogical models of the upper mantle, through the interpretation of seismic velocity models. The composition of natural orthopyroxenes (Pbca space group) can be represented, to a first approximation, as lying along the enstatite (Mg$_2$Si$_2$O$_6$)-ferrosilite (Fe$_2$Si$_2$O$_6$)-diopsid line (Frisillo et al. 1976). A single-crystal X-ray refinement of the Pbca space group produced unit-cell parameters of $a = 18.268$ Å, $b = 8.836$ Å, $c = 5.196$ Å, and unit-cell volume $V = 883.7$ Å$^3$. The corresponding density at 1 bar and 298 K is 3.209 g/cm$^3$.

From these earlier studies, the substitution of Fe for Mg in the Opx structure appears to decrease slightly the bulk modulus and more significantly the shear modulus. The influence of minor element substitutions has been less investigated; though such data are required to fully constrain the elastic properties of natural upper mantle pyroxenes. Chai et al. (1997) determined the elastic constants of a natural Opx from Kilbourne Hole by impulsive stimulated light scattering, and report bulk and shear moduli substantially higher than those of pure orthoenstatite.

Static compression experiments by Hugh-Jones and Angel (1997) support this conclusion, and show that substitution of even small amounts of Al and Ca in the orthoenstatite structure causes an increase in the bulk modulus by as much as 14%.

However, from these previous studies it is difficult to discriminate the effects of the different cations, and only a net behavior from all the minor cations can be described. To isolate the effect of Ca-substitution on the elastic behavior of Opx, we performed Brillouin scattering measurements of sound velocities and elastic moduli on synthetic single crystals of Ca-bearing orthoenstatite.

EXPERIMENTAL PROCEDURES

Starting material

The crystals used in this study were synthetic Ca$_{0.07}$Mg$_{1.93}$Si$_2$O$_6$ orthopyroxenes. These samples come from the same synthesis product described by Nestola and Tribaudino (2003), and have been accurately characterized by means of electron microprobe analysis, transmission electron microscopy, and X-ray diffraction (Nestola and Tribaudino 2003). A single-crystal X-ray refinement in the Pbca space group produced unit-cell parameters of $a = 18.268$ (2) Å, $b = 8.836$ (1) Å, $c = 5.196$ (1) Å, and unit-cell volume $V = 883.7$ (2) Å$^3$. The corresponding density at 1 bar and 298 K is 3.209 g/cm$^3$.

Three single crystals of high optical quality and free of inclusions were pre-orientated with faces nearly normal to either the $a$, $b$, or $c$ orthorhombic axes. They were ground and polished with two parallel sides to 100×150 μm platelet-shaped samples with thickness of 30–40 μm. The crystals were then glued to glass fibers, mounted on goniometer heads, and transferred to a three-circle Eulerian cradle that was used to control the sample orientation on the Brillouin system.

Brillouin measurements

The Brillouin spectra were collected using a six-pass tandem Fabry-Perot interferometer and an argon laser ($\lambda_c = 514.5$ nm) as the light source. The scattered light was detected by a solid-state photon detector. The incident laser power was 100 mW and collection times for a spectrum were generally 5–10 min. All measurements were performed with a platelet symmetric scattering geometry and a 90° scattering angle ($\theta$). This geometry allows the determination of the acoustic velocities independent of the refractive index of the sample (Withfield et al. 1976) or nearly so. Further experimental details are given in Sinogeikin et al. (2004). The portion of the light that was inelastically scattered by acoustic phonons was used to...