A RAMAN SPECTROSCOPIC STUDY OF Fe-Mg OLIVINES

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End-member synthetic fayalite and forsterite and a natural solid-solution composition crystal of composition $(Mg_{1.80},Fe_{0.20})SiO_4$ as well as polycrystalline forsterite and fayalite isotopically enriched in ^{26}Mg and ^{57}Fe, respectively, were synthesized and their Raman spectra were measured. The observed isotopic shift of vibrational modes permits the assignment of spectra, especially in low-frequency region, to be improved. The low wavenumber Raman modes in olivine are best described as lattice modes consisting to a large degree of mixed vibrations of M(2) cation translations and external vibrations of the SiO_4 tetrahedra. The polarized single-crystal spectra of forsterite and $Fo_{90}Fa_{10}$ were recorded at a number of temperatures from room temperature to about 1200 °C. From these data, the microscopic Grüneisen parameters for three different A_g modes for both compositions were calculated, and also the structural state of the solid solution crystal was investigated. The difference in the measured mode wavenumbers between the heating and cooling is due to hysteresis effect. The spectra of the $Fo_{90}Fa_{10}$ crystal, unlike the spectra of forsterite, show a discontinuity in the wavenumber behavior for the mode at ~220 cm^{-1} at 700-1000 °C upon heating and cooling (Fig. 1). Both these observations are discussed taking into account the various crystalline effects, i.e. annealing of the point defects, structural transformation, rigid rotation of the SiO_4 tetrahedra, changing of the oxygen-oxygen interaction. The discontinuity in the wavenumber behavior of the mixed $Mg/T(SiO_4)$ mode (Fig. 1) may be related to variations in the Fe-Mg intracrystalline partitioning behavior in the $Fo_{90}Fa_{10}$ crystal, i.e. to some decrease in the concentration of Fe^{2+} at M(2).
The mode wavenumber and intensity behavior of internal SiO_4 vibrations as a function of temperature are discussed in terms of crystal field and dynamic splitting and also ν_1 and ν_3 coupling. Crystal field splitting increases very slightly with temperature, whereas dynamical field splitting is temperature dependent. The degree of ν_1-ν_3 coupling decreases with increasing temperature.

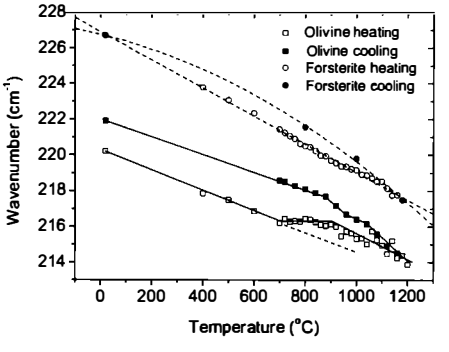

Fig. 1. Temperature dependence of the wavenumber of the A_g mode at 220 cm^{-1} in Fo_{100} and $Fo_{90}Fa_{10}$.