Reflectance measurements of mid- and far-infrared spectroscopy for crystalline olivine

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Abstract

We have measured reflectance for crystalline olivine ($(Mg_{92}Fe_8)_2SiO_4$). The measurements were conducted in 2007 and 2008 with FT-IR spectrometer (Bruker IFS120HRX) at BL43IR beam line of SPring-8. Temperature range was 300, 250, 200, 150, 100, and 60 K and wavelength range is 2000-100 cm^{-1} (5-100 micron). Reflection spectra were measured for each surface by using polarized infrared beam parallel to a, b, and c axes because crystal structure of olivine was the orthorhombic which had distinct three surfaces. When the temperature decreases, we confirmed the spectral features that; shift to shorter wavelength, increase intensity, and become sharp. Olivine crystal belongs to the space group Pnma (D^{16}_{2h}), and there are 35 IR active optical modes (9 91u, 13 B2u, 13 B3u) expected from symmetry analysis. We have observed 8 in the axis c (B1u), 12 in the axis b (B2u), and 11 in the axis a (B3u) features. The dielectric constants of the crystal have also derived by fitting the reflection spectra with dispersion formula of damped oscillators. Our measured spectra of crystalline olivine are useful for analysis of astronomical studies of low temperature environment.

Introduction

Crystalline silicate materials such as olivine and pyroxene were confirmed in comets, circumstellar environment, and young stellar objects based on the spectral identifications from ISO and ground-based telescope infrared observations (e.g., Waelkens et al. 1996; Waters et al. 1996; 1998; Malfait et al. 1998; Molster et al. 1999; Honda et al. 2003). Laboratory works were conducted to investigate the temperature dependence of crystalline silicates and the absorption spectra of silicate particles was measure in low temperatures (e.g., Koike et al. 2006) because simple spectral fits suggests that much of the optically thin silicate emissions occurs at temperatures in 50-100 K range (e.g., Bowey et al 2002).

Absorption measurement of powder samples in laboratory is a conventional way to look into the thermal behavior of the infrared features of solids, but the absorption spectra by powder depend on the shape distribution, the size distribution and the coagulation degree of samples, which are difficult to be regulated. It is also difficult to know which optical axis an absorption feature belongs to. On the other hand, if we are able to obtain a single crystalline sample with good quality and reasonable size, its reflection measurements at infrared region provide with little ambiguity the dielectric or optical constants of crystal's each optical axis, which is vital for accounting the radiation characters of crystalline dust.

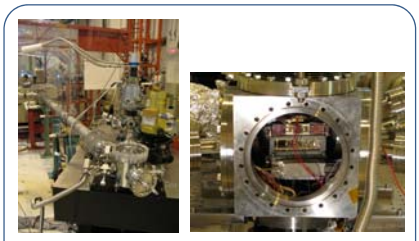

Figure 1.
Optical systems of FT-IR spectrometer in Spring-8. A sample stage were installed in a chamber.

Measurement

Reflectance for crystalline olivine ($(Mg_{92}Fe_8)_2SiO_4$) were measured with FT-IR spectrometer (Bruker IFS120HRX) at BL43IR beam line of SPring-8 in 2007 and 2008 (Figure 1). Ohmic heaters and silicon diode temperature sensors were on the sample stage in the cryostat, and the sample temperature was controlled by a Lakeshore temperature controller. The cryostat was kept vacuum with pressure less than 10^{-4} Torr. Spectral resolutions 1cm^{-1} was used, and 0.5, 0.25, and 0.125 cm^{-1} when we needed to resolve narrow features.

Olivine single crystal was produced in Kohistan (Figure 2). Three pieces of a few millimeter size crystal were prepared from a large single crystal, and crystalline axes were determined by the X-ray precession method. Three pieces were polished to make the (100), (010), and (001) surfaces.

Reflection spectra for the infrared beam polarized parallel to the axis a was measured on (100) surface, the axis b on both (100) and (001), and the axis c on both (100) and (010). Infrared spectra have been obtained at the temperature of 300, 200, 100, and 50 K using a reflectance module of 10 degree incident angle. A beam splitter and wire grid polarizer in the PE substrate (~400-50 cm^{-1}) and Thallium Bromo-Iodide (KRS5) substrate (2000-~400) were employed. A customized optical module was used to derive the spectrometer beam to and from the sample compartment through the window on the cryostat wall. The reflectance of the sample was calibrated with a gold-coated silicon substrate mirror that was also on the sample stage and inserted to the spectrometer beam path in the sample way of the olivine sample. 100 percent reflectance for all wavelength was assumed for the gold mirror.

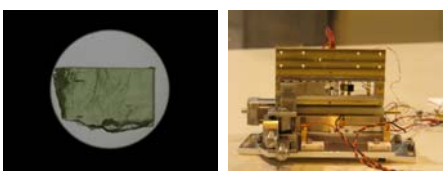

Figure 2.
Measured millimeter size sample of the clistalline olivine and the sample stage.

Results

Figure 3 shows the measured reflectance for crystalline olivine of each axis as a function of wave number. When the temperature decreased from 300 to 60 K, the intensity of reflectance changes drastic in linger wavelength (<~700 cm^{-1}). Olivine crystal belongs to the space group Pnma (D^{16}_{2h}) and there are 35 IR active optical modes (9 91u, 13 B2u, 13 B3u) expected from symmetry analysis (Ishii 1978). 8 in the axis c (B1u), 12 in the axis b (B2u), and 11 in the axis a (B3u) features have been clearly observed in our measurements.

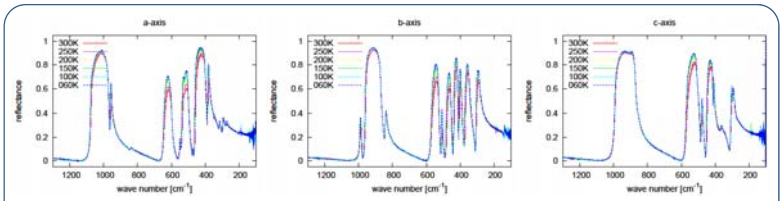

Figure 3.
Reflectances for crystalline olivine of the a axis (left), b axis (center) , and c axis (right) as a function of wave number [cm^{-1}]. The colors show the differences of the temperatures for the crystal.

Discussion

The Dielectric constants are derived from the reflection spectra by adopting the classical dispersion model of dumped oscillator (Bohren and Huffman 1983). The dispersion relation is expressed as,

$$R(\omega) = \left| \frac{\sqrt{\epsilon(\omega) - \sin^2\theta} - \cos\theta}{\sqrt{\epsilon(\omega) - \sin^2\theta} + \cos\theta} \right|^2$$

where ε_0 is the dielectric constant at the short wavelength limit, ω_{pj}^2, ω_j^2, and γ_j are the oscillator strength, transverse optical wavelength, and dumping factor, respectively. The reflectance R is calculated from $\varepsilon(\omega)$,

$$\epsilon(\omega) = \varepsilon_0 + \sum_j \frac{\omega_{pj}^2}{\omega_j^2 - \omega^2 - i\gamma_j\omega}$$

where ϑ is the beam incident angle to a surface of the crystal (10 degree). We use the least-square method to fit the parameters from reflectance. The results of the oscillator parameters are shown in Table.

Table.
Oscillator parameters for each axis at 300K. We assumed 11, 12, and 8 features for a, b, and c axes respectively from results of the reflectances.

a-axis (B3u): ε_0=2.77			b-axis (B2u): ε_0=2.70			c-axis (B1u): ε_0=2.71		
ω_i	ω^2_{pi}/ω^2_i	γ_i/ω_i	ω_i	ω^2_{pi}/ω^2_i	γ_i/ω_i	ω_i	ω^2_{pi}/ω^2_i	γ_i/ω_i
9.764E+02	6.718E+00	3.131E+05	9.842E+02	5.059E+00	7.006E+03	8.741E+02	5.383E+00	4.681E+05
9.580E+02	3.649E+00	1.486E+05	8.718E+02	2.794E+00	2.921E+05	5.029E+02	9.920E+00	1.097E+05
8.409E+02	8.427E+00	2.850E+03	8.378E+02	9.471E+00	9.026E+04	4.762E+02	8.033E+00	3.457E+04
6.040E+02	1.261E+01	8.008E+04	5.269E+02	1.250E+01	4.914E+04	4.151E+02	5.624E+00	1.614E+05
5.017E+02	1.284E+01	8.753E+04	5.060E+02	6.315E+00	1.120E+04	4.084E+02	5.645E+00	8.116E+04
4.023E+02	5.593E+00	2.473E+05	4.559E+02	8.902E+00	3.883E+04	3.045E+02	4.644E+00	2.680E+03
3.792E+02	5.511E+00	1.410E+05	4.168E+02	5.410E+00	4.806E+04	2.911E+02	2.951E+00	1.046E+05
3.196E+02	2.785E+00	8.486E+03	3.951E+02	5.337E+00	7.208E+04	2.761E+02	2.627E+00	6.119E+03
2.942E+02	3.454E+00	2.808E+04	3.487E+02	5.488E+00	1.471E+05			
2.748E+02	2.694E+00	3.982E+03	2.891E+02	4.421E+00	1.361E+05			
2.009E+02	2.459E+00	1.029E+03	2.768E+02	2.528E+00	6.413E+03			
			1.434E+02	2.078E+00	1.603E+03			

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