The optical properties of olivines: bulk optical constants and absorption spectra of small particles

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Abstract

The optical constants of crystalline silicates (forsterite and Mg-rich olivine) are determined from the reflectance measurements of bulk samples. The calculations using the bulk optical constants show that the absorption spectra are highly dependent on the particle shapes. The calculated spectra of Rayleigh particles are compared with the spectra measured in the laboratory.

1 Introduction

The emission features in the mid and far-infrared spectra observed by the Infrared Space Observatory have given the clear evidence of the presence of the crystalline silicates in a variety of astronomical objects. The crystalline silicates are identified as Mg-rich olivines or pyroxenes from the comparison between the observed features and the absorption spectra of the powdered samples measured in the laboratories.

However, in the laboratory, the powdered samples are usually embedded in the matrix of KBr or polyethylene; therefore the measured spectrum is not identical to that of the grains in vacuum. Furthermore, the different conditions of preparing sample pellets easily induce the different spectra, because the absorption spectrum can be affected by the size and shape distributions of the grains as well as the degree of dispersion in the matrix.

On the other hand, the optical constants of bulk crystal samples provide the more definite and reliable standards for the study of crystalline dust, since the optical constant is independent on a sample as long as the sample has an identical component. The absorption spectra of particles can be calculated by assuming the size and shape distributions of particles.

We have measured the bulk optical constants of synthesized forsterite Mg_2SiO_4 (hereafter, Fo100) and natural Mg-rich olivine $(Mg_xFe_{1-x})_2SiO_4$ of which Mg/(Mg+Fe) number ratio x is 0.9138 (hereafter, Fo91). In this paper, we present the results of the reflectance measurements and compare the laboratory spectra of small particles with the spectra calculated by using the bulk optical constants.

2 Reflectance measurements and bulk optical constants

The forsterite (Fo100) single crystal synthesized by the Czochralski method (Takei and Morioka, 1974) was provided by Dr. Tsuchiyama. The olivine (Fo91) is a natural sample from St. John's Island in Egypt. The crystalline axes of bulk samples were determined by X-ray precession method. Three pieces for each sample were cut perpendicular to the crystalline axes and their surfaces have been polished.

Reflection spectra have been measured at the incident angle of 10° using a Nihonbunko Fourier spectrometer. The electric field of incident light have been polarized along the crystal axis and perpendicular to the plane of incidence by using the wire-grid polarizer with substrate of KRS5 and polyethylene. The frequency resolutions are $4\mathrm{cm}^{-1}$ in the range of wavelength $\lambda = 7$ to 20 μm and $1\mathrm{cm}^{-1}$ in $\lambda = 20$ to 40 μm.

The spectra have been analyzed by classical dispersion theory and have been fitted to the multiple oscillators model by using the Marquardt method (Marquardt 1963). The reflectivities for the three different crystallographic axes of Fo100 and Fo91 are shown in Figure 1. The derived parameters of oscillators and the detailed discussion including the comparison with the previous works by Servoin & Pirious (1973) and Steyer (1974) will be provided by Suto et al. (*in preparation*).

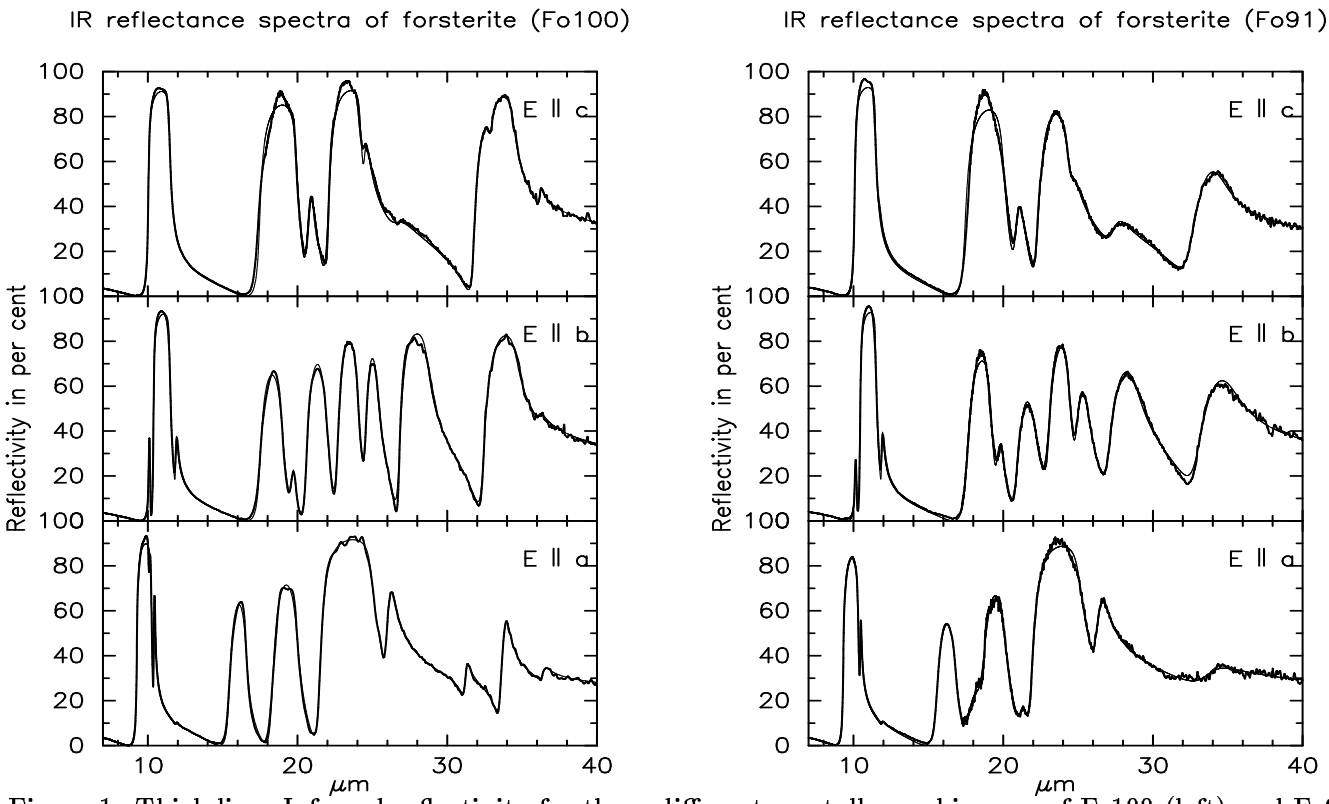

Figure 1. Thick line: Infrared reflectivity for three different crystallographic axes of Fo100 (left) and Fo91 (right). Thin line: The best fit calculated by using the classical oscillator model.

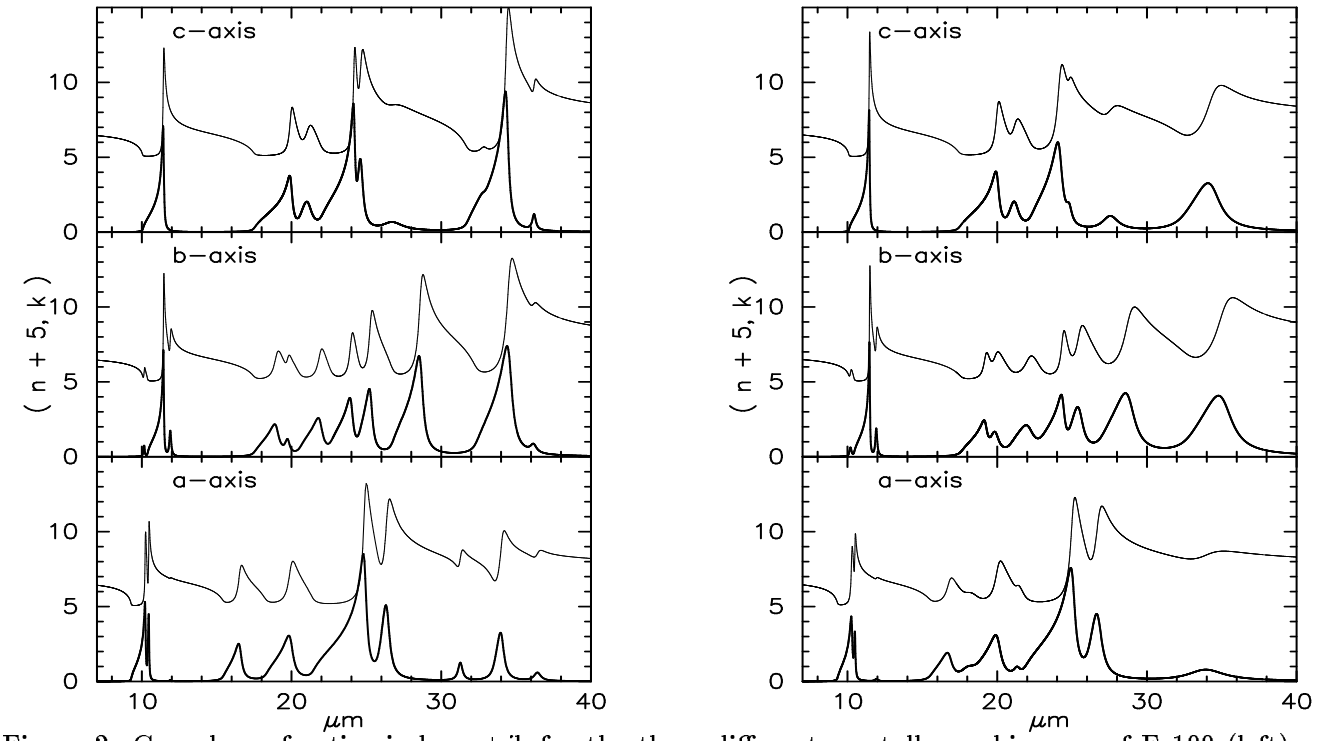

Figure 2. Complex refractive index n+ik for the three different crystallographic axes of Fo100 (left) and Fo91 (right). Thin line: n+5. Thick line: k.

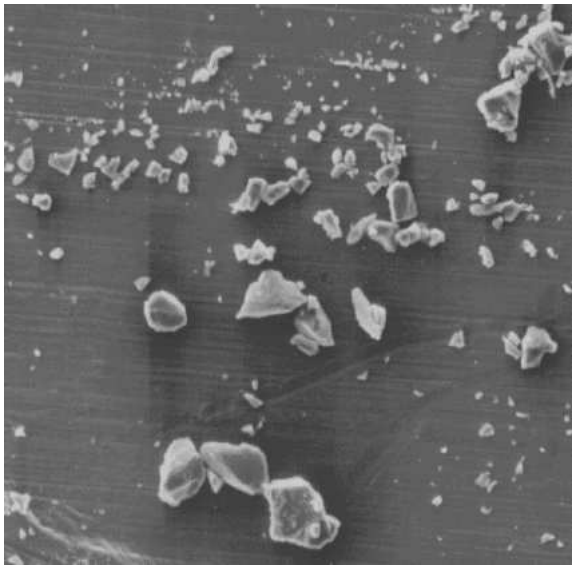

Figure 3. A scanning electron microscope image of Fo100 particles.

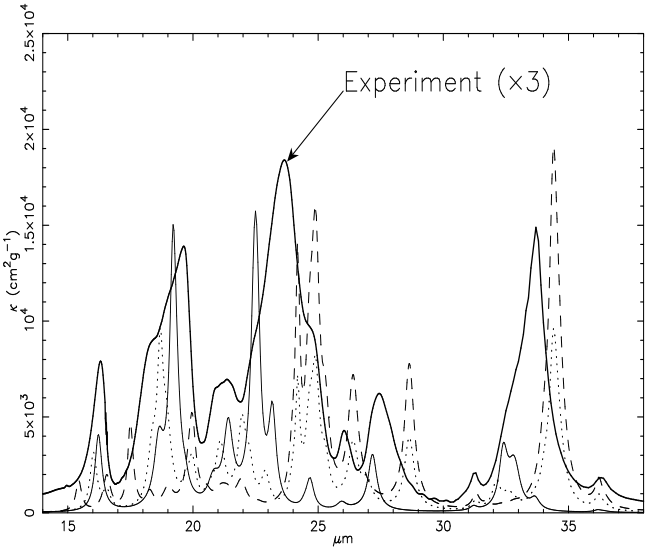

Figure 4. Absorption spectra calculated from the optical constants of bulk Fo100 sample for spheres (thin), randomly oriented needles (dotted), and randomly oriented disks (broken), compared with the MAC spectra of Fo100 particles multiplied by 3 (thick).

3 Absorption spectra of small particles

Powdered samples of Fo100 were produced by grinding the bulk crystal for transmittance measurements. Figure 3 shows the scanning electron microscope (SEM) image of Fo100 particles. The powder is embedded in a KBr matrix for the spectroscopy in the wavelength shorter than 25μm and in a polyethylene sheet for the spectroscopy in the wavelength longer than 25μm. The size of the particles in KBr are less than $\sim$0.3μm and particles in polyethylene are smaller than $\sim$1μm.

Comparison of the mass absorption coefficients (MAC) of Fo100 obtained from the transmittance measurements with the absorption spectra calculated from the bulk optical constants for various shapes are shown in Figure 4. In the calculations, Rayleigh approximation is assumed. It can be seen that the shape of the particles has striking effect on the positions and intensities of the prominent resonance features. The profiles and peak strengths of the features are quite different from the laboratory data of small particles.

Since the SEM images show various shapes of particles, we consider a continuous distribution of ellipsoids (CDE; Bohren & Huffman 1983) as an approximation of shape distribution; the CDE model consists of various shapes of triaxial ellipsoids including spheres, needles, and disks. The spectra calculated for a CDE compared with the laboratory spectra of particles are shown in Figure 5. In Figure 5b, the MAC spectrum of a natural $(Mg_{0.9}Fe_{0.1})_2SiO_4$ sample from Akita in Japan is drawn as a representative of the Fo91 laboratory spectrum. There is a very good coincidence between the calculated spectrum for the CDE model and the measured spectrum. Figure 6 demonstrates the matrix effects of KBr/polyethylene for a CDE. Although the intensities of the features are different, there is no significant change in the peak positions. On the contrary, if the shape of particles is assumed to be sphere, there are shifts of peak positions up to $\sim$0.6 μm.

Because the emission features observed in the spectra of the astronomical objects have been identified with the crystalline silicate features by comparison of the observed spectra with the laboratory data of particles, the close resemblance between the CDE calculation and the laboratory spectra suggests that crystalline silicate (forsterite and olivine) grains around these astronomical sources are nonspherical.

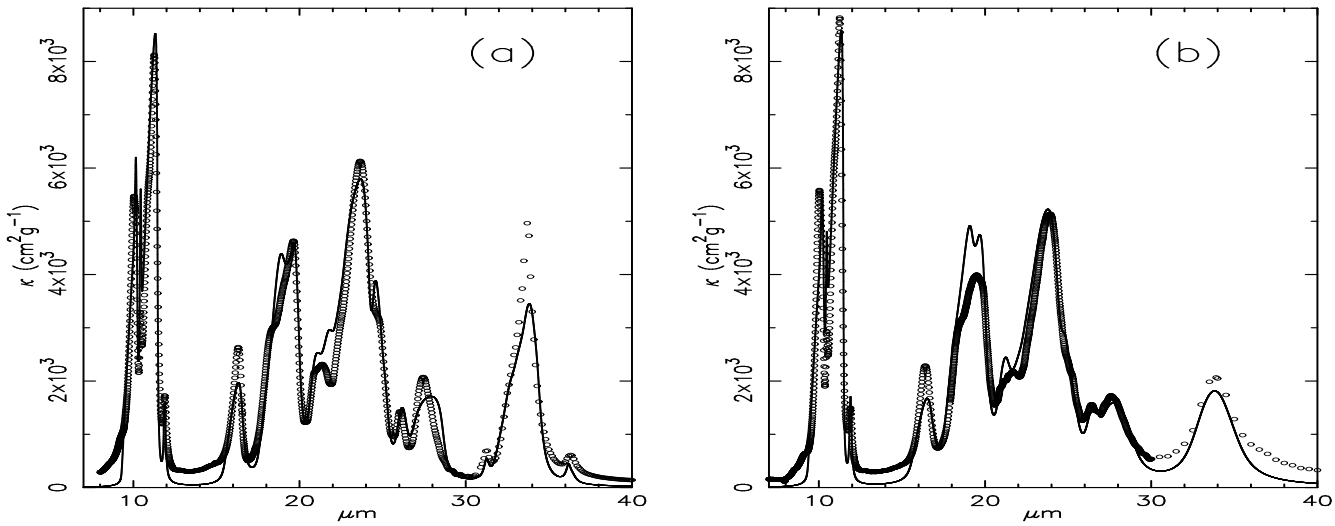

Figure 5. (a) The comparison of the absorption spectrum calculated from the optical constants of bulk Fo100 for a CDE (solid line) with the MAC spectrum measured in the laboratory for Fo100 particles (circles). (b) Same as (a) except that the bulk optical constants are those of Fo91 and MAC are measured for $(Mg_{0.9}Fe_{0.1})_2SiO_4$ particles.

4 Summary

The optical constants of crystalline silicates (forsterite and Mg-rich olivine) are determined from the reflectance measurements of bulk samples.

The main results of the calculations with the bulk optical constants are as follows.

- Absorption spectrum is largely dependent on the shape of particles.
- The laboratory spectrum of small particles is similar to the calculated spectrum for a CDE.
- For the CDE model, the peak positions of absorption features are not so changed by the matrix effects of KBr/polyethylene.

The similarity between the laboratory spectra and the calculated spectra for CDE suggests that the crystalline silicate grains around the astronomical objects are nonspherical.

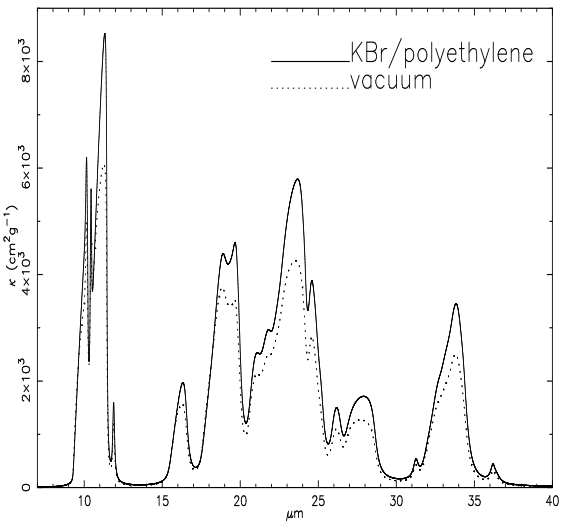

Figure 6. Comparison of the calculated MAC of a CDE in KBr/polyethylene and in vacuum from the optical constants of Fo100.

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