Constraints on cosmic silicates

V. Ossenkopf1,2, Th. Henning1,2, and J.S. Mathis3

1 Universitäts-Sternwarte, Schillergäßchen 2, O-6900 Jena, Federal Republic of Germany
2 Max-Planck-Gesellschaft, Arbeitsgruppe “Staub in Sternentstehungsgebieten”, c/o Universitäts-Sternwarte, Schillergäßchen 2, O-6900 Jena, Federal Republic of Germany
3 Washburn Observatory, University of Wisconsin, 475 N. Charter Street, Madison WI 53706-1582, USA

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Abstract. Observational determinations of opacities of circumstellar silicates, relative to the peak value near 10 μm, are used to estimate the optical constants n and k, the real and imaginary parts of the index of refraction. Our model assumes that the observed opacities represent grains that have a power-law size distribution, \( n(a) \propto a^{-3.5} \), and a continuous distribution of ellipsoidal shapes. The absolute absorption per volume of grains, \( C_{abs}(\lambda)/V \), is constrained by laboratory measurements of the static dielectric constants of silicate minerals.

Circumstellar dust is modified by processing within the interstellar medium. This leads to higher band strengths and a somewhat larger ratio of the opacities at the 18 and 10 μm peaks, compared with circumstellar silicates.

By using an effective-medium theory, we calculate the effects of small spherical inclusions of various materials (various oxides, sulfides, carbides, amorphous carbon, and metallic iron) upon silicate opacities. Some of these can increase the absorption coefficient k in the 2–8 μm appreciably, as is needed to reconcile laboratory silicate opacities with observations of both the interstellar medium and envelopes around late-type stars. Others inclusions affect the 18-μm-to-10-μm band ratio.

We give tables of two sets of optical constants for warm oxygen-deficient and cool oxygen-rich silicates, representative for circumstellar and interstellar silicates. The required opacity in the 2–8 μm region is provided by iron and magnetite.

Key words: circumstellar matter – dust, extinction – infrared: interstellar – continuum – radio continuum: interstellar

1. Introduction

For the solution of various astrophysical problems, the accurate knowledge of the optical data of cosmic dust grains over a broad wavelength region is required. The optical constants of interstellar graphite and silicate grains by Draine & Lee (1984, hereafter DL), tabulated in Draine (1985), give a good representation of the opacities of grains in the diffuse interstellar medium (ISM) over the entire wavelength range over which the opacities can be estimated from observations. The DL constants are based on a combination of laboratory measurements and astronomical observations. It would be very desirable to identify the DL "astrophysical silicate" in terms of real materials. Recently, there has been considerable progress in laboratory measurements of cosmic dust candidates (Bussoletti et al. 1988), the analysis of circumstellar and interstellar dust emission (Allamandola & Tielens 1989), and the analysis of IRAS data from many objects with circumstellar dust.

Cosmic grains are quite different in various environments, such as circumstellar regions, the ISM of various densities (ranging from the diffuse ISM to the dense cores of molecular clouds), or in protostellar disks. Even within a particular type of environment there are individual variations, such as among the silicate bands in various M-star spectra, or in the unidentified infrared emission band strengths in the diffuse ISM.

In this paper, we discuss the opacities and optical constants of silicates. From the observed opacity of circumstellar dust, relative to the value at 10 μm, we estimate optical constants with the assumption of a size and shape distribution. We then use laboratory data to produce a model of the optical constants of dirty silicates. We try to relate observationally-based and laboratory-based constants to each other. Finally, we discuss changes to the optical constants that take place within the diffuse ISM.

We will refer to the 0.8 – 5 μm region as the near-infrared (NIR); to the 5 – 40 μm, containing the major silicate absorption features near 10 and 18 μm, as the mid-infrared (MIR), and the 40 – 400 μm region as the far-infrared (FIR).

2. Methods for the derivation of optical constants

From either astronomical observations or laboratory measurements one has empirical determinations of the extinction or absorption of materials as a function of wavelength. From these data the complex dielectric constant \( \varepsilon = \varepsilon' + i\varepsilon'' \) must be derived, comprising two functions of wavelength through its real and imaginary parts. The second relation connecting \( \varepsilon(\lambda)' \) and \( \varepsilon(\lambda)'' \) is either of two conditions derived from causality: the Kramers-Kronig relations, or else a dispersion model given by the Clausius-Mosotti law for dielectrics (see Bohren & Huffman 1983).

It is often convenient to express the complex dielectric function, \( \varepsilon \), by the complex index of refraction, \( n \), where \( \varepsilon = n^2 \). We will use \( n \) and \( k \), where \( m = n + ik \).

One method applicable for both astronomical and laboratory determinations of optical constants of solids is transmission spectroscopy of fine particles. In the laboratory the material is usually
in the form of very small grains, embedded in a transparent matrix material that has a known dielectric constant, $\varepsilon_m$. Usually the extinction by the grains is interpreted by assuming that each particle is a sphere (of radius $a$), that each is in the Rayleigh limit ($a \ll \lambda/2\pi$), that the grain material is isotropic, and there is no clumping of the particles. With these assumptions, the scattering is small and the extinction is the same as the absorption. The measurements provide values of $C_{abs}(\lambda)/V$, the total absorption cross section of grains per unit volume of grains. In the Rayleigh limit, $C_{abs}(\lambda)/V$ is independent of $a$. Then

$$\frac{C_{ext}(\lambda)}{V} = \frac{C_{abs}(\lambda)}{V} = \frac{6\pi}{\lambda_m} \text{Im} \left( \frac{\varepsilon(\lambda) - 1}{\varepsilon(\lambda) + 2} \right),$$

(1)

where $\varepsilon = \varepsilon_{grain}/\varepsilon_m$ is the dielectric function of the particles relative to the embedding matrix, $\lambda$ is the wavelength in vacuum, and $\lambda_m = \lambda/\lambda_m$. If the imaginary part of the function $f(\lambda) = (\varepsilon(\lambda) - 1)/(\varepsilon(\lambda) + 2)$ is known, we can derive $\text{Re}[f(\lambda)]$ from either the Kramers-Kronig relations or a Clausius-Mosotti dispersion analysis, which includes a least-squares fit to the absorption by Lorentzian profiles (see an extensive discussion by Dorschner et al. 1988). A disadvantage of the dispersion analysis is the fine-tuning of the fit, which may overlook small features of the spectrum.

Martin & Rouleau (1991) pointed out, and we independently realized, that the Kramers-Kronig relations apply to the function $f(\lambda)$ as well as to $\varepsilon$ itself. Then

$$\text{Re}[f(\lambda)] = \frac{2}{\pi} P \int_0^\infty \frac{\lambda^2}{\lambda^2 - \Lambda^2} \text{Im}[f(\Lambda)] \, d\Lambda.$$

(2)

Here $P$ indicates that the principal value of the integral has to be taken. Some uncertainty results from the fact that $\text{Im}[f(\lambda)]$ is known only over a finite wavelength interval and must be extrapolated for the computation of the integral. The extrapolation is usually accomplished by assuming power laws, the exponents of which depend on the structure of the grains (e.g. amorphous vs. crystalline: Tielens & Allamandola 1987). The contributions from the very high frequency end may be summed up in a real parameter $\varepsilon_\infty$ which leads to a systematic shift in the absolute values of $\text{Re}[f(\lambda)]$. With this information we can derive the complete complex function, $f(\lambda)$, from which the dielectric function $\varepsilon$ can be calculated directly. Rouleau & Martin (1991) recently used this method, modified to account for deviations from spherical shapes, to the analysis of laboratory carbon data.

A theoretical check whether a given set of opacities $C_{abs}(\lambda)/V$ is physically possible comes from the Kramers-Kronig relation and the condition that the static real dielectric function of every material has to be positive, leading to the constraint

$$\frac{1}{3\pi^2} \int_{\lambda_1}^{\lambda_2} \frac{C_{abs}(\lambda)}{V} \, d\lambda < 1,$$

(3)

(Purcell 1969). The quantity $n_m$ is the real refractive index of the surrounding medium. The limits of the integral are arbitrary wavelengths between 0 and $\infty$. The inequality applies to an arbitrary size distribution of spherical grains. Equation (3) shows that the absorption has to fall more steeply than $\lambda^{-1}$ at very long wavelengths to enable convergence of the integral for $\lambda_2 \rightarrow \infty$.

### 3. Application to astronomical silicates

#### 3.1. Absorption efficiencies of silicates in circumstellar shells

Infrared emission from the circumstellar dust shells of late-type stars provides strong constraints on the optical properties of the dust. However, the emission is mainly influenced by the distribution of temperature and density within the circumstellar shell. Even if the dust shell is optically thin, it is impossible to extract the opacity of the dust from the emitted flux alone without modelling the physical conditions in the dust shell (Henning et al. 1983; Bedijn 1987). If the shell is optically thick, a full radiative transfer treatment is needed (see Gürtler et al. 1989 for a discussion). Angular size measurements of dust emission should provide important additional constraints on the physical parameters of the dust and shell (Harvey et al. 1991; Collision & Fix 1991).

The IRAS low-resolution-spectrograph (LRS) data show a rather wide variety in shape of the emission profiles in the 8–23 $\mu$m region (Vardya et al. 1986; Cheeseman et al. 1989; Onaka et al. 1989; Simpson 1991). The possible origin of these differences was considered by Nuth & Hecht (1990). We will mainly discuss the general behaviour of the silicate features, including the ratio of the two silicate absorption peaks, $b = C_{abs}(18 \mu$m)/$C_{abs}(10 \mu$m). We also consider the position and width of the 10 $\mu$m feature. The maximum of the bending vibration has a rather constant position at about 18 $\mu$m, but the width is quite uncertain (4–10 $\mu$m).

Figure 1 shows the silicate opacities as derived by various authors; Table 1 lists some of their properties. In Table 1, all arguments are wavelengths in $\mu$m and the arguments 10 and 18 refer to the respective maxima. The table also gives the absolute strength $C_{abs}(10)/V$, the wavelength of its peak, and the $C_{abs}(\lambda)/C_{abs}(10)$ ratios for various wavelengths.

![Fig. 1. Observationally determined opacities for circumstellar silicate dust derived by Bedijn (1987, dotted line), Volk & Kwok (1988, short-dashed line), and David & Papoular (1990, long-dashed line). The solid line represents the opacities produced by an MRN-size distribution of silicates from Draine & Lee (1984).](image)

Bedijn (1987) represented NIR and IRAS observations of M-type Miras and OH/IR stars with a parametrized opacity mainly based upon the spectrum of OH 26.5+0.6. Volk & Kwok (1988, hereafter VK) used IRAS data of 467 AGB stars to give a mean opacity function. The David & Papoular (1990, hereafter DP) values are derived from the spectra of two M-type supergiants.
Table 1. Characteristic parameters of observational opacity functions

<table>
<thead>
<tr>
<th></th>
<th>(C_{\text{abs}}(10)/V) (\left[\text{m}^{-3}\right])</th>
<th>Peak pos. (\text{FWHM}) (\left[\mu\text{m}\right])</th>
<th>(C_{\text{abs}}(18)/C_{\text{abs}}(10))</th>
<th>(C_{\text{abs}}(2)/C_{\text{abs}}(10))</th>
<th>(C_{\text{abs}}(100)/C_{\text{abs}}(10))</th>
<th>(C_{\text{abs}}(200)/C_{\text{abs}}(10))</th>
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<td>DL</td>
<td>1.0</td>
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<td>2.7</td>
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<td>0.13</td>
<td>0.010</td>
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<td>-</td>
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<td>2.4</td>
<td>0.41</td>
<td>0.47</td>
<td>0.017</td>
</tr>
<tr>
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<td>-</td>
<td>10.0</td>
<td>1.8</td>
<td>0.28</td>
<td>0.073</td>
<td>-</td>
</tr>
<tr>
<td>Bedijn</td>
<td>-</td>
<td>9.5</td>
<td>3.2</td>
<td>0.50</td>
<td>0.47</td>
<td>0.045</td>
</tr>
<tr>
<td>Simpson</td>
<td>-</td>
<td>9.7–10.2</td>
<td>2.6–2.8</td>
<td>0.40</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Simpson (1991) used the LRS spectra of 117 late-type stars whose silicate emission features appear to be optically thin. The plotted values in the UV should be considered with caution, since they are extrapolated from the NIR observations without taking into account the optical activity of silicates due to electronic transitions. Therefore, we will not derive optical constants from the opacities for this wavelength range. For comparison, we list the DL opacity, based upon the Orion Trapezium emission for the 10\(\mu\text{m}\) region and the 18-\(\mu\text{m}\) observations of oxygen-rich stars and the dust absorption towards the Galactic center. We did not use the opacity of Rowan-Robinson (1986), since it is too large at long wavelengths because of a wrong calibration of IRAS data at 100\(\mu\text{m}\).

We stress that the band ratio \(b\) is dependent upon the model used for the circumstellar dust. DL and Simpson (1991) assumed a value of \(b\). The value discussed by most authors lies between 0.3 and 0.5. Bedijn (1987) found that OH/IR stars exhibit a higher \(b\) value up to 0.8, while Miras stars have lower \(b\) of about 0.5. This was explained by cooler dust grains in the envelopes of OH/IR stars. The most comprehensive investigation of \(b\) was made by VK, who found a mean value of 0.41. The peak position of the 10\(\mu\text{m}\) feature lies in the interval 9.5–10.2\(\mu\text{m}\), and the FWHM in the range 1.8–3.2\(\mu\text{m}\).

The absorptivity of silicates in the NIR is important for the energy balance in circumstellar envelopes because the stellar radiation field peaks in that region. Typical terrestrial silicates have only very weak absorption in this region, with \(k < 0.001\). However, Jones & Merill (1976) and Bedijn (1977) have shown that a simulation of the IR fluxes of late-type M stars is only possible if the silicate grains exhibit a considerable absorption in the NIR, and there is similar evidence from polarimetric observations (Ravenhead 1991). Jones & Merrill proposed “dirty silicates”, with an optical behaviour derived from meteoritic rocks (Dorschner 1968). This silicate dust contains nickel-iron material. Various authors’ estimates of \(k\) for dirty silicates differ by an order of magnitude. At 2\(\mu\text{m}\) Rogers et al. (1983) suggested \(k \approx\) 0.01, while Jones & Merrill (1976) found 0.1. DL took a mean value of 0.03. Collison & Fix (1991) found \(Q_{\text{abs}}/Q_{\text{esc}} \approx 5\) which results from the DL optical constants and a grain radius of \(a = 0.1\mu\text{m}\) to be too low to explain their high-resolution maps of circumstellar emission. Their implication was that the optical properties of the circumstellar silicates must be different than those of DL. One reason could be a too low \(k\) value, although a different size distribution cannot be excluded. It seems reasonable to assume that 0.03 \(\leq k \leq 0.1\). We will produce a model (Sect. 7) with \(k = 0.07\).

We think it likely that such large values of \(k\) are produced by mixtures of other materials into the silicates, rather than by color centers alone (because they are associated with discrete NIR features that are not observed) or by damage from cosmic rays.

There is also considerable uncertainty concerning the behaviour in the FIR. In Sect. 2 we saw that \((C_{\text{abs}}(\lambda)/V)\text{ must decrease more steeply than }\lambda^{-1}\text{ for very long wavelengths. Solid-state models assume there a power-law behaviour }C_{\text{abs}}(\lambda)/V\propto \lambda^{a}\text{. In case of layer-lattice silicates }n\approx 1–1.5\text{ and for 3-dimensional crystalline silicates }n\approx 2\text{ is expected (Tielens & Allamandola 1987). Figure 1 shows that the DL and VK curves exhibit a final }\lambda = 2\text{ behaviour at }100\mu\text{m, in contrast to the other two functions which show a flatter decrease because of an upturn between 40 and }60\mu\text{m. Sopka et al. (1988) estimated an independent absolute value. Taken at }\lambda = 200\mu\text{m it corresponds to }C_{\text{abs}}(200)/C_{\text{abs}}(10) = 0.01–0.02\text{. The observational determination of the FIR opacity must come from the satellite ISO and from sub-mm observations.}

3.2. Derivation of optical constants from the opacities

We will derive optical constants from the observed circumstellar silicate opacities, \((C_{\text{abs}}(\lambda)/V)\text{, by using the Kramers-Kronig relations or a dispersion model as described in Sect. 2, but relaxing the assumption that the grains are spherical and always small enough to be in the Rayleigh limit. In contrast to the laboratory, the grains are better isolated and the “embedding” material is practically a vacuum.}

While the grains are almost surely small in comparison to infrared wavelengths, the situation for optical radiation is not so clear. Unfortunately, there is little information on the particle size distribution in stellar dust shells. Bedijn (1987) discussed grain sizes of about 0.1\(\mu\text{m}\); DP assumed values between 0.05 and 0.1\(\mu\text{m}\). Polarization measurements by Ravenhead (1991) suggest particle sizes of \(a \lesssim 0.1\mu\text{m}\). We will assume the MRN (Mathis et al. 1977) power-law size distribution, with \(n(a) \propto a^{-3.5}\) in the interval 0.01\(\mu\text{m} < a < 0.25\mu\text{m}\), since it contains sizes within the ranges likely to be present in circumstellar dust.

There is no reason to believe that circumstellar grains are spherical. We considered a continuous distribution of ellipsoids (CDE) with random orientations. Ellipsoids of the same volume may be characterized by two parameters, \(L_1\) and \(L_2\) depending on the axial ratios (Bohren & Huffman 1983). The distribution among shapes is given by an assumed probability \(P(L_1, L_2)\). Bohren & Huffman (1983) consider the simplest, \(P(L_1, L_2) = \text{constant}\). Rouelle & Martin (1991) applied this probability in the analysis of laboratory data and found that the absorption spectra were influenced at long wavelengths (\(\lambda > 20\mu\text{m}\)) only.

We will assume here a distribution

\[
P(L_1, L_2) = 120 L_1 L_2 L_3
\]
where $L_3 = 1 - L_1 - L_2$.

This distribution has the physically reasonable properties of being a maximum for spheres ($L_1 = L_2 = 1/3$) and being zero for the extreme values $L_1$, $L_2$ or $L_3 = 0$ (infinitely thin needles or flattened pancakes).

We used an iterative correction method, described in the appendix, to take the size and shape distribution into account. This method corrects the optical constants so that the $C_{\text{abs}}(\lambda)/V$ calculated for the nonspherical and large grains agree with the observations.

In general, the optical constants determined by the Kramers-Kronig relations and those found by Lorentzian profile fitting agree extremely well, but the Clausius-Mosotti approach smooths details of features. This is not necessarily a disadvantage; for instance, the unphysical discontinuities in slope of the DP absorption efficiencies at 8 and 60 $\mu$m shown in Fig. 1 are repeated in the $k$ function derived by the Kramers-Kronig approach but are smoothed in the Clausius-Mosotti analysis. In the following, we use the Kramers-Kronig because it is a more direct method.

Two additional parameters required in the derivation procedure are the electronic contribution $\varepsilon_\infty$ to the dielectric function and the absolute value of the absorption. We choose $\varepsilon_\infty$ from laboratory data of silicates. The published values of $\varepsilon_\infty$ for different silicate glasses lie in the rather narrow range between 2.7 and 3.4 for olivines and pyroxenes (Dorschner 1988). We will select the same mean value of 2.94, as in DL. The exact magnitude is not critical for the further calculations.

Observations provide only relative values of $[C_{\text{abs}}(\lambda)/V]_{\text{obs}}$. We will normalize to the maximum value of $[C_{\text{abs}}(\lambda)/V]_{\text{obs}}$ in the 10 $\mu$m range. There is a wide range of measured data for laboratory silicates, with values between 0.2 $\mu$m$^{-1}$ (Stephens et al. 1988, amorphous olivine smoke) and 2.3 $\mu$m$^{-1}$ (Koike et al. 1989, crystalline obsidian), although some of the extreme cases may be due to errors in measurement. The large spread may be caused by structural differences, since crystalline silicates show, in general, higher values than their amorphous counterparts.

We estimate $C_{\text{abs}}(10)/V$ from the static dielectric constant, $\varepsilon_0$, which the Kramers-Kronig relations directly relate to the strengths of all resonances. If $\varepsilon_\infty$ and $C_{\text{abs}}(\lambda)/C_{\text{abs}}(10)$ are known for $\lambda < \lambda_0$, where $\lambda_0$ is in the FIR, and no further resonances occur for $\lambda > \lambda_0$, $\varepsilon_0$ is directly related to $C_{\text{abs}}(10)/V$. If there are some further resonances for $\lambda > \lambda_0$, only an upper limit to the $C_{\text{abs}}(10)/V$ can be set. To demonstrate the relation between the absolute value of absorption and the long wavelength limit of the dielectric function, we show in Fig. 2 the optical constants calculated via the Kramers-Kronig analysis from the relative absorption efficiencies given by DP with a normalization constant of 0.68 $\mu$m$^{-1}$, 0.90 $\mu$m$^{-1}$ and 1.13 $\mu$m$^{-1}$. These correspond to $\varepsilon_0 \approx 5.0$, 6.1, and 7.6, respectively.

The $\varepsilon_0$ of various glasses is between 3.8 for pure quartz glass and 16 for lead glass (Landolt-Börnstein 1957). Mg silicates show values between 5.5 and 6.5; Fe silicates between 6.8 and 7.5 (Landolt-Börnstein 1957; Touloukian & Ho 1981). We use an average value of 6.5, or a limit of the refractive index of $\varepsilon_0^{1/2} = 2.55$. For the DP data, this $\varepsilon_0$ leads to a $C_{\text{abs}}(10)/V$ of 0.98 $\mu$m$^{-1}$, assuming that no further resonances occur at longer wavelengths. For both the Bedijn and VK data this condition leads to $C_{\text{abs}}(10)/V = 0.57 \mu$m$^{-1}$.

In Fig. 3 we compare the optical constants obtained from the relative opacities of the different authors; for a particular opacity of Bedijn (1987; $b = 0.5$, $C_{\text{abs}}(100)/C_{\text{abs}}(10) = 0.045$) and
for those from VK and DP, and also those of Draine (1985). Figure 3 shows that the different absorption efficiencies in the NIR are translated into $k$ values between 0.015 and 0.07. We see that the optical constants derived from the observational opacities are relatively similar. In the $n$ values only the DL constants deviate at long wavelengths (because of their choice of $e_0$). In the $k$ behaviour the differences in the 10–30 $\mu$m region due to the different selection of the band behaviour are more obvious. The turnover at longer wavelengths to a $k^{-1}$ or $k^{-1/4}$ behaviour in the Bedijn or DP opacities, respectively, lead to a discontinuity in the slope of the $k$-values that seems unphysical. The Bedijn constants seem unreasonable at these long wavelengths.

3.3. Observations of the silicate component in the interstellar dust

The extinction and emission properties of the dust in the diffuse ISM do not allow a straightforward derivation of opacities for silicate grains because there is dust from various sources present. Interstellar grains may well have silicates heavily mixed with other materials, such as forms of solid carbon or carbides (e.g., Mathis & Whittet 1989), so their optical properties cannot be applied directly to silicates alone. IRAS observations require rather high emission in the 12 and 25 $\mu$m filters, but carbon grains, inhomogeneous silicate-carbon grains or dark mantles on the grains, as well as dirty silicates, can contribute significantly to the emission. A separate silicate opacity function derived from observations of the diffuse ISM may not exist. However, the diffuse ISM provides observational constraints on the nature of the material producing the infrared silicate features.

The shape of the 10-$\mu$m interstellar silicate absorption feature appears to be similar to circumstellar emission profiles. However, for the ISM $b \approx 0.6$ (Roche 1988), in contrast to the circumstellar value of 0.41, so there is some processing of the silicates within the ISM. Of course, the $b$ is not determined by the silicates alone, and the admixture of other materials into the silicates can change it appreciably (see Sect. 6).

To determine $C_{\text{abs}}(10)/V$ in the diffuse ISM, one needs to observe the optical depth of the silicate feature, $\tau(10)$, through the diffuse ISM towards objects with no silicate features in their own spectra. This is a very difficult task.

The ratio $A_V/\tau(10)$ for the diffuse ISM is estimated to be about 8 towards the Galactic centre (Becklin et al. 1978) and 40, with a considerable uncertainty, towards WC stars (Cohen et al. 1989). For the same stars Roche & Aitken (1984) had found a value of 18; the discrepancy arises from the presence of strong emission in the stellar atmospheres from the 7.7 $\mu$m unidentified emission bands. From depletion ratios (Whittet 1984) one can estimate that the contribution of the silicates to the total visual extinction has to be considerable, perhaps between 60% and 80%. The difference between the local value of $A_V/\tau(10)$ and that towards the Galactic center might be the result of a different Si/C ratio in the dust associated with the gradient in (O/H) (Wink et al. 1983; Shaver et al. 1983) that may cause that the contribution of silicates to $A_V$ be larger in the inner parts of the Galaxy. Of course, the ratio $A_V/\tau(10)$ is sensitive to the size distribution in addition to composition.

It turns out that the visual extinction of an MRN size distribution of grains with a refractive index in the range typical for silicates is nearly independent of the uncertain absorption coefficient $k$. An enhanced $k$ does not produce a higher extinction at visual wavelengths ($C_{\text{ext}}(0.55)/V_{\text{MRN}} \approx 6.4 \mu$m$^{-1}$ for silicates). Consequently, the $A_V/\tau(10)$ ratio constrains only $C_{\text{abs}}(10)/V$. Taking the metallicity effect into account we obtain $C_{\text{abs}}/V$ peak values between 1.0 $\mu$m$^{-1}$ and 0.3 $\mu$m$^{-1}$. Polarization measurements by Aitken (1989) suggest band strengths between 1.4 $\mu$m$^{-1}$ and 0.75 $\mu$m$^{-1}$.

The FIR opacity is very uncertain for the diffuse ISM. If we convert the absorption values given by Hildebrand (1983), Renegarajan (1984), Fujita et al. (1989), and Casey (1991) with their power laws to a common wavelength of 200 $\mu$m we obtain relative absorption efficiencies of $C_{\text{abs}}(200\mu m)/V = 0.004 – 0.02 \mu$m$^{-1}$. In our final model we come out with a value for silicates near the lower limit, suggesting that other components may also contribute considerably.

4. Constraints to the structure of astronomical silicates

Both the smooth structure of the silicate band profiles and condensation calculations indicate that circumstellar and interstellar silicate dust must be highly amorphous. Cosmic abundances make Mg and Fe silicates, such as amorphous olivines and pyroxenes, preferred.

Nuth & Hecht (1990) suggest on the basis of laboratory data that the silicates in rapidly cooling circumstellar flows are highly oxygen deficient, while the silicates in the ISM and slowly cooling flows exhibit a stoichiometric Si/O ratio known from chemical-equilibrium silicates. This suggestion is consistent with an empirical ordering of the silicate emission in Mira variables (Stencel et al. 1990), based on the IRAS Low-Resolution Spectrometer spectra of Little-Marenin & Little (1990). O-deficient silicates have a lower peak opacity, a lower $b$, and a lower NIR absorptivity. Included Fe may be reduced by Si to form metallic inclusions, while in more O-rich compositions Fe probably appears as oxides such as magnetite (Fe$_3$O$_4$) and hematite (Fe$_2$O$_3$). Temperature effects (Day 1976) suggest that the absolute band strengths and the band ratio $b$ should be larger in the cold ISM than in circumstellar dust. The formation of mantles in the diffuse ISM and in envelopes around late-type stars is still an open issue. The existence of such mantles might also influence band ratio and large parts of the absorption spectrum.

Various condensation calculations suggest that the silicate grains in oxygen-rich atmospheres may condense on cores or with inclusions of some other materials. Such materials as MgO (Frideyev 1988), MgS (Köhler 1991), Al$_2$O$_3$ (Onaka et al. 1989), metallic Fe, Fe$_3$O$_4$, and Fe$_2$O$_3$ (Laureijis 1989; Nuth & Hecht 1990) have been proposed.

5. Optical constants of laboratory silicates

There are a number of investigations of optical constants of terrestrial silicates (cf. Dorschner & Henning 1986). Unfortunately, they are mostly crystalline. We know of only a few studies of the dielectric functions of amorphous silicates over a broad wavelength region including the mid-infrared. Studies of some optical properties of oxygen-deficient silicates (Wada et al. 1991; see also Nuth & Donn 1982) do not yet allow a direct derivation of optical constants, but did confirm the theoretical prediction of Nuth & Hecht (1990) that O-deficient silicates show a much lower band ratio $b$.

In Fig. 4 some laboratory-based optical constants of amorphous silicate are shown, along with the relative absorption efficiencies $C_{\text{abs}}(1)/V$ for the Rayleigh limit. Figure 4a contains the curves for pyroxenes ((Mg,Fe)$_2$SiO$_4$), Fig. 4b for olivines ((Mg,Fe)$_2$SiO$_4$). The data come from Day (1979, 1981),
Fig. 4. Optical constants and opacities for Rayleigh grains of (a) laboratory pyroxenes: bronzite (Dorschner et al. 1988, solid lines), FeSiO$_3$ (Day 1981, dotted lines), and MgSiO$_3$ (Day 1979, 1981, dashed lines) and (b) laboratory olivines: irradiated olivine (Kr"atschmer & Huffman 1979, solid lines), Fe$_2$SiO$_4$ (Day 1981, dotted lines), and Mg$_2$SiO$_4$ (Day 1979, 1981, dashed lines).

<table>
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<th>$C_{\text{abs}(10)}/V$</th>
<th>Peak pos.</th>
<th>FWHM</th>
<th>$C_{\text{abs}}(18)/C_{\text{abs}(10)}$</th>
<th>$C_{\text{abs}}(2)/C_{\text{abs}(10)}$</th>
<th>$C_{\text{abs}}(100)/C_{\text{abs}(10)}$</th>
<th>$C_{\text{abs}}(200)/C_{\text{abs}(10)}$</th>
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<td>9.6</td>
<td>2.3</td>
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<td>1.7 $10^{-3}$</td>
<td>0.011</td>
<td>2.7 $10^{-3}$</td>
</tr>
<tr>
<td>FeSiO$_3$</td>
<td>0.71</td>
<td>9.5</td>
<td>1.6</td>
<td>0.41</td>
<td>-</td>
<td>0.028</td>
<td>0.014</td>
</tr>
<tr>
<td>MgSiO$_3$</td>
<td>0.96</td>
<td>9.6</td>
<td>2.3</td>
<td>0.28</td>
<td>-</td>
<td>6.8 $10^{-3}$</td>
<td>2.9 $10^{-3}$</td>
</tr>
<tr>
<td>Fe$_2$SiO$_4$</td>
<td>0.23</td>
<td>9.8</td>
<td>1.9</td>
<td>0.89</td>
<td>-</td>
<td>0.043</td>
<td>0.020</td>
</tr>
<tr>
<td>Mg$_2$SiO$_4$</td>
<td>0.73</td>
<td>9.9</td>
<td>2.1</td>
<td>0.36</td>
<td>-</td>
<td>0.014</td>
<td>4.3 $10^{-3}$</td>
</tr>
<tr>
<td>irr. olivine</td>
<td>1.42</td>
<td>9.8</td>
<td>1.5</td>
<td>0.36</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 2. Characteristic parameters of the opacities of amorphous laboratory silicates.
Krätzschmer & Huffman (1979), and Dorschner et al. (1988). Instead of the original curves from Dorschner et al. (1988) we used updated measurements for bronzite (MgO,FeO,SiO2) carried out in polyethylene and corrected for clumping effects. They should be more accurate at long wavelengths. However, the measurement of optical constants is difficult, and the published values may contain still errors; for instance, the refractive index of bronzite at long wavelengths seems high. We could show that fractal clumping (Ossenkopf 1991) or size effects are not responsible for the high values.

From Fig. 4a we see that the positions of the absorption maxima and shapes of the 10-μm features are rather similar for various pyroxenes, but that > 20 μm the absolute values of the optical constants differ. The contribution of the large strength of the 18-μm feature of bronzite to the Kramers-Kronig integral results in a higher refractive index at long wavelengths than for other silicates. Figure 4b shows that the olivine curves have similar 10-μm features but deviate in the 20-μm region. The Day (1979, 1981) curves, except the FeSiO3, show the bending vibration at longer wavelengths (about 20 μm) than the others and the observations. In general, the 10-μm feature of pyroxenes peaks at shorter wavelengths than in case of olivines.

Table 2 lists the widths, central positions, and some characteristic opacity ratios of small (a < 0.1 μm) spherical particles of the various amorphous laboratory silicates, similar to Table 1 for the opacities of astronomical silicates. The FeSiO3 sample and the irradiated olivine differ from the other laboratory silicates as well as from the observationally-based parameters (see Table 1). The observations demand a rather broad 10-μm feature. This is best fulfilled by the bronzite sample, MgSiO3, and MgFeSiO3. These silicates have Cabs(10)/V = 0.75 – 1.0 μm-1, b = 0.3 – 0.5, and Cabs(100)/Cabs(10) = 0.01 – 0.03.

6. Effects of inclusions

We now consider the effects on the optical constants of small spherical inclusions of the materials proposed in Sect. 4. We used the Maxwell-Garnett mixing rule for volume fractions of the inclusions up to 5% and the Bruggeman mixing rule for values between 5 and 20% (see Ossenkopf 1991). The application of effective-medium theories at short wavelengths requires that all inclusions are very small but still large enough that optical bulk constants are valid. We calculated Cabs/V for the resulting inhomogeneous grains, applying the Rayleigh approximation up to the shortest wavelengths in order to demonstrate the changes in the optical constants without being influenced by size effects. References for the used optical constants of the inclusions are compiled in Table 3.

In Fig. 5 we show the effects of small dielectric inclusions (Al2O3, MgO, and MgS) in bronzite. The volume fraction of 10% of the inclusions is taken from Onaka et al. (1989). The inclusions have almost no effect in the NIR and FIR. The inherent absorption bands of the admixed dielectrics in the MIR only change the appearance of the silicate bands. The Al2O3 shifts the maximum position of the 10-μm feature to 9.7 μm and increases its FWHM to 2.7 μm; the MgO admixture increases the band ratio b to 0.68. All other parameters remain the same as for pure bronzite. MgS causes no changes in the characteristic parameters.

Figure 6 shows the results for 3% Fe, 6% magnetite (Fe3O4), and 6% hematite (Fe2O3) inclusions. These volume fractions, chosen to include the same number of Fe atoms, arise if we assume that all olivines and pyroxenes (with a continuous range in the Fe/Mg ratio) have the same probability of occurring. Figure 6 shows a dramatic increase in the absorption efficiency in the NIR and a very small change of the feature shapes. Magnetite inclusions produce the highest NIR absorption. The Cabs(2)/Cabs(10) value is increased to 0.04 for Fe, 0.08 for Fe3O4, and 0.22 for Fe2O3. The k-values at 2 μm are 0.009 for Fe, 0.014 for Fe3O4, and 0.05 for Fe2O3. Such inclusions could explain the dirtiness of silicates. Note that there is no essential enhancement of the absorption in the FIR by the inclusions. Only Fe2O3 produces an enhanced band ratio (b = 0.64). The real part of the refractive index is systematically increased for all three types of inclusions relative to the pure bronzite, by about 0.08 at 2 μm and by about 0.20 at 200 μm.

For both Al2O3 and Fe2O3 we had only optical constants of the α-type structure available. Although very small isolated particles of these media tend to change into the γ-structure, we will assume that the α-structure is preserved due to the embedding into the silicate matrix material.

In the diffuse ISM the silicate grains are mixed with grains
from carbon stars, and composite grains may form (e.g., Jones et al. 1990 and ref. therein; Mathis & Whiffen 1989). Also the accretion of carbon and other refractory elements from the gas phase should result in mantles on the silicates. The admixture of carbon strongly affects the absorption of silicates. In Fig. 7 we show $C_{abs}(\lambda) / V$ for bronzezite grains with inclusions of amorphous carbon and with a mantle of the same material, both taking 5% of the grain volume. Optical constants for amorphous carbon were taken from Edoh (1983) because they represent a material with a rather high number of graphic bonds, as should appear in the diffuse ISM. Carbon leads to a similar enhancement of the absorption coefficient in the NIR as the iron-rich inclusions. $C_{abs}(2)/C_{abs}(10)$ is increased to 0.17 for inclusions and 0.27 for mantles. The silicate bands are hardly influenced.

![Figure 7](image1.png)

Fig. 7. Opacities for Rayleigh grains of pure bronzezite (solid line), bronzezite with 5% inclusions of amorphous carbon (dotted line), and bronzezite grains with a mantle of amorphous carbon with the same volume fraction (dashed line).

In the FIR, carbon as a mantle absorbs much more effectively than as an inclusion. Carbon mantles produce a $C_{abs}(200)/C_{abs}(10)$ of 0.010, compared to 0.003 for inclusions. Both absorption values are still in the observational range (see Sect. 3.3). This mantle effect is strongly dependent on the optical constants of the materials considered; other kinds of amorphous carbon, with lower values of the optical constants in the FIR (e.g. Rouleau & Martin 1991), produce much lower differences between inclusions and mantles.

Another important inclusion might be SiC. In Fig. 8 we show the absorption in the region of the silicate bands for bronzezite grains with inclusions of 2%, 5%, 10%, and 15% SiC (optical constants from Palik 1985). Up to 10% by volume, the SiC feature can be hidden within the 10-μm silicate band. This might explain why the SiC feature is not observed in the ISM (Whittet et al. 1990).

![Figure 8](image2.png)

Fig. 8. Opacities for Rayleigh grains of pure bronzezite (solid line) and bronzezite with inclusions of silicon carbide: 2% (dotted line), 5% (short-dashed line), 10% (long-dashed line), and 15% (very-long-dashed line).

SiC is a material with strongly negative Re[$\varepsilon(\lambda)$] in the region of the 11.2-μm feature, similar to graphite near 0.22 μm. Free-standing grains of such materials can have a strong resonance for surface modes, but the central wavelength and breadth of the resonance are influenced by the shapes of the grains. We investigated whether deviations from spheres can broaden the
11.2-μm feature of SiC inclusions enough to hide it within the silicate band. Figure 9 shows the results for 15% SiC inclusions as an extreme CDE (i.e., \( P(L_1, L_2) = \text{constant} \)), and also as spherical inclusions. We see that for inclusions shape effects are relatively small. We estimate that about 10% SiC is an upper limit to the amount of SiC which can occur in grains in the diffuse ISM without the 11.2-μm feature becoming obvious in absorption. The 10% upper limit on SiC inclusions in the diffuse ISM is consistent with the 20% measured in IRC+10216 (Martin & Rogers 1987) if we assume that about half of interstellar grains are contributed by carbon-rich stars (Jura & Kleinmann 1990). The percentage may be still lower since the study of grain evolution processes indicates that a considerable amount of the interstellar grain mass must have been contributed from molecular clouds where the formation of SiC particles seems to be unlikely.

For all of the mixtures considered here (even in the case of the SiC admixture) the resulting optical constants are still relatively similar to the pure bronzite and show no negative dielectric functions so that strong surface resonances and shape effects are not expected for the resulting inhomogeneous grains. Nevertheless, shape effects can slightly enhance the band ratio \( b \) and the FIR absorption.

### 7. Optical constants for astronomical silicates

In the following, we will present two sets of optical constants. One should be typical for warm oxygen-deficient circumstellar silicate dust, in which silicates have not reached their stoichiometric proportions (as suggested by Nuth & Hecht 1990). The second set is for cool oxygen-rich dust representative for shells of OH/IR stars and the diffuse ISM. There is clearly a range of circumstellar dust properties, especially of the profile of the 10-μm band (Stencel et al. 1990), so our derived constants are simply representative.

We selected the Volk & Kwok values (see Fig. 3) to represent warm slightly oxygen-deficient circumstellar dust because VK used a very careful derivation procedure, the data represent a broad sample of stars, and the results seem physically reasonable. Because of a lack of detailed observational data in the NIR, all observationally derived opacities seem rather unreliable in the region 2–7.5 μm. Therefore, we replaced the VK opacities in this wavelength range by those calculated for laboratory silicates with iron and iron oxide inclusions. The effective optical constants of this mixture at these wavelengths are nearly independent of the type of silicate, all of which have very small \( k \) and similar values of \( n \). The volume ratios of the inclusions were chosen to produce the same \( C_{\text{abs}}(2)/V \) as VK. The two opacity sets were joined smoothly between 7 and 8 μm. The band ratio of 0.41 indicates that the silicates, and therefore iron, are only partially oxidized, so we chose equal numbers of Fe atoms in metallic iron and magnetite. If there are dust shells with still shorter cooling times or higher dust temperatures than the typical VK star, \( b \) can be still lower and Fe might be present as pure iron.

The static value of the dielectric function for our silicate mixed with Fe and FeO inclusions will be different from that of a pure silicate, used in Sect. 3.2. Changing \( \epsilon_0 \) leads to another normalization \( C_{\text{abs}}(10)/V \) of the opacities. Therefore, an iteration procedure similar to that described in the appendix is necessary to produce a model with inclusions for which the \( [C_{\text{abs}}(2)/C_{\text{abs}}(10)], C_{\text{abs}}(10)/V \), and the static dielectric function are consistent with each other. The resulting model has 6.3% FeO, 3.1% Fe, \( C_{\text{abs}}(10)/V = 0.64 \mu m^{-1} \), and an enhancement of \( \epsilon_0 \) by 1.9 relative to that of pure silicate. The opacity \( \kappa(10) = C_{\text{abs}}(10)/\rho V = 2.010^8 \text{cm}^2 \text{g}^{-1} (\rho \text{ is the bulk density of silicates: } 2.8–3.3 \text{g cm}^{-3}) \).

Another problem with these inhomogeneous grains is the application of the Kramers-Kronig relation for the derivation of optical constants in combination with effective-medium “theories”. The Bruggeman mixing rule does not conserve the Kramers-Kronig invariance. Test calculations show, however, that for such grains with inclusions as described above the rule yields only very small deviations from the Kramers-Kronig behaviour. Therefore, a combination of the mixing rules with the Kramers-Kronig analysis seems justified. The resulting set of optical constants is given in Fig. 10.

For the cool oxygen-rich silicates we do not have observational opacities directly. We introduced two artificial changes into the VK absorption efficiencies to give them the behaviour we expect in oxidized grains (Nuth & Hecht 1990), in which the silicates contain a stochiometric oxygen content; a higher value of the peak absorption, and a higher band ratio. Iron inclusions may attract some oxygen, and the ratio between pure iron and iron oxide is lowered. We simulate this effect by a ratio of 0.1 between the numbers of Fe atoms in metallic iron and magnetite. This produces some minor changes in the NIR behaviour. The absolute value of the peak absorption is increased to \( C_{\text{abs}}(10)/V = 0.75 \mu m^{-1} \), or \( \kappa(10) = 2.510^8 \text{cm}^2 \text{g}^{-1} \). To produce the enhanced band ratio of about 0.6 the 18-μm band was taken to be that of bronzite with iron and magnetite inclusions. A smooth transition to the VK curve at longer wavelengths was introduced between 25 and 35 μm. The resulting optical constants are also given in Fig. 10. Table 4 contains the numerical \( n \) and \( k \) values for the two silicate materials.

It is clear that interstellar grains might be strongly modified by chemical and physical processing after their expulsion from stellar atmospheres. If half of dust is contributed by C-rich stars, about half of the Si, Fe, and other elements in grains should be originally in carbides, or at least in forms other than oxides and silicates. However, almost all Si must be in silicates to account for the strength of the silicate features per H atom, as has been emphasized by Tie lens & Allamandola (1987).

We can calculate the number of Si atoms per H atom needed to produce \( A_V/\tau(9.7) = 40 \) with our optical con-
Table 4. Optical constants for warm O-deficient circumstellar silicates (Set 1) and cool O-rich interstellar silicates (Set 2)

<table>
<thead>
<tr>
<th>λ [μm]</th>
<th>n</th>
<th>k</th>
<th>λ [μm]</th>
<th>n</th>
<th>k</th>
</tr>
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<tr>
<td>4.000-1</td>
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<td>7.712-2</td>
<td>1.160-1</td>
<td>2.105</td>
<td>7.684-1</td>
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<tr>
<td>7.692-1</td>
<td>1.850</td>
<td>5.731-2</td>
<td>1.200-1</td>
<td>2.208</td>
<td>6.132-1</td>
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<td>1.000+0</td>
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<td>5.462-2</td>
<td>1.220-1</td>
<td>2.200</td>
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<td>1.250+0</td>
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<td>2.214</td>
<td>5.175-1</td>
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<td>1.492+0</td>
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<td>7.289-2</td>
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<td>4.679-1</td>
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<tr>
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<td>7.333-2</td>
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<td>2.500+0</td>
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<td>7.500+0</td>
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<td>7.887-1</td>
<td>4.922</td>
<td>9.131-1</td>
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Constants. Observationally, the visual colour excess per H column density is $E(B-V)/N(H) = 1.710^{-22}$ mag cm$^2$ (H atom)$^{-1}$ (Bohlin et al. 1978) for the diffuse ISM. The ratio $A_V/E(B-V) = 3.1$, and our absorption cross section per Si atom, derived above, is $[C_{abs}(9.7)/V]μ/ρ$, where $μ$ is the average mass of the silicate molecules producing the observed bands (110–120 atomic mass units). Thus we have to require the abundance ratio of Si in silicates $[N(Si)/N(H)]_{silicate}$ to be $3.1×[A_V/E(B-V)]/E(B-V)/N(H)](μ/ρ)^{-1}$, or $3.2×10^{-5}$, which is the entire cosmic abundance of Si.

Our constants are able to provide the necessary absorption at 9.7 μm only if there has been an almost complete oxidation of the Si, Fe, etc. from C-stars into silicates within the ISM (Whittet et al. 1990), although some SiC appears as inclusions in meteorites (Zinner et al. 1991). These oxidation processes probably prevent metallic Fe from being present in the ISM (Jones 1990).

8. Conclusions

We could show that meaningful optical constants for cosmic silicates can be derived from infrared observations. The optical constants were interpreted in terms of the optical behaviour of inhomogeneous grains calculated on the basis of laboratory data.

The following results were obtained:

(i) We have seen that measurements of the opacities of circumstellar dust provide relative opacities $C_{abs}(λ)/C_{abs}(10)$, that $C_{abs}(2)/V$ is much larger than for laboratory silicates, and that the NIR emissivity of interstellar dust shows that its absorption in the NIR is at least as great, or greater, than that of circumstellar dust surrounding M stars. It is possible to explain the observed dirtiness of silicates in the NIR by inclusions of absorptive, especially iron containing materials.

(ii) Silicates are modified within the ISM by processing and mixing with dust from carbon stars. The general optical properties of circumstellar silicates and the interstellar silicate component are relatively similar. The value of $b$ for interstellar dust, 0.6 (Roche...
not in the Rayleigh limit. The size distribution does not enter the $C_{\text{abs}}(\lambda)/V$ at long wavelengths because all grains are small compared to the wavelength, in which case $C_{\text{abs}}(\lambda)/V$ is independent of size for a given shape.

In general, $C_{\text{abs}}(\lambda)/V^{(i)}$ is larger than $[C_{\text{abs}}(\lambda)/V]_{\text{obs}}$ in the optical because large grains are somewhat more efficient absorbers per volume than small (until $a > \lambda$), and small grains were used to estimate the trial constants ($n^{(i)}, k^{(i)}$) from which $C_{\text{abs}}(\lambda)/V^{(i)}$ was determined. For longer wavelengths, the $C_{\text{abs}}(\lambda)/V^{(i)}$ is larger than $[C_{\text{abs}}(\lambda)/V]_{\text{obs}}$ because the CDE distribution contains shapes that are better absorbers than the spheres. Also the band ratio $b$ is slightly enhanced.

Let $g^{(i)}(\lambda) = C_{\text{abs}}(\lambda)/V^{(i)}/[C_{\text{abs}}(\lambda)/V]_{\text{obs}}$. Our reasoning is that if the $n, k$ from Rayleigh-sized spheres produce, with larger sizes in a CDE, an absorption that is $g^{(i)}$ too large, we should analyze the observed absorption reduced by a factor of $g^{(i)}$. So, we obtain a corrected $C_{\text{abs}}(\lambda)/V$ spectrum $[C_{\text{abs}}(\lambda)/V]_{\text{obs}}/g^{(i)}$. Our second iteration of trial constants, $n^{(2)}, k^{(2)}$, is found by substituting this corrected spectrum in place of the $[C_{\text{abs}}(\lambda)/V]_{\text{obs}}$ and estimating a $C_{\text{abs}}(\lambda)/V^{(2)}$ as we did for $C_{\text{abs}}(\lambda)/V^{(i)}$. This procedure was continued for 3 or 4 iterations, until the $C_{\text{abs}}(\lambda)/V$ calculated using the MRN and CDE distributions matched the observed one.

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