

Laboratory Astrophysics of Circumstellar Dust

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Abstract. This paper summarizes laboratory investigations of stardust analogue materials with special emphasis on the spectroscopic behaviour of silicates and carbonaceous materials. The review will also deal with the identification of the observed features in the dust spectra of AGB stars.

1. Introduction

Small solid particles form an important component of the circumstellar (CS) environment of AGB stars. Although they are only minor contributors to the total mass of the envelopes, they are of decisive importance for their thermal, dynamical, and chemical state. IR and submm/mm continuum radiation is produced by the thermalization of stellar light. The gas is heated through collisions between dust particles and the gas atoms and molecules. The momentum in the outflows is supplied by radiation pressure acting on dust grains and the collisional transfer of momentum to the gas. The surfaces of dust grains can act as catalytic sites for the formation of molecules. In the outer regions of the envelopes, dust grains play an important role for shielding the molecules from dissociative interstellar UV radiation and cosmic rays. AGB stars are the main sources of freshly nucleated interstellar grains. Therefore, these objects are important for the chemical evolution of grain populations in galaxies, whose properties influence the birth of the next generation of stars.

A deeper understanding of the complex interplay between hydrodynamics, radiation transfer, and chemistry has to be based on a better knowledge of the microphysics of CS dust. Guided by IR spectroscopy, submm/mm observations, and the analysis of stardust in primitive meteorites, laboratory astrophysics must contribute to the understanding of dust formation from the gas phase, the formation of molecules by surface reactions, and the optical behaviour of small solid particles. On the other hand, AGB star envelopes are interesting regions for the study of grain formation and molecular chemistry under rather extreme conditions, usually difficult to reproduce in terrestrial laboratories.

The review will mainly deal with the optical properties of stardust analogue materials. Without better optical data, the radiative transfer modelling of spectral energy distributions and the interpretation of absorption and emission features observed in the dust spectra of AGB stars will remain ambiguous.

For a more detailed discussion of dust properties and evolution in our galaxy, the interested reader is referred to recent reviews of this topic (Dorschner & Henning 1996; Henning 1997, 1998a, 1998b; Sandford 1996).

2. Stardust materials - An overview

A key parameter for dust formation in the outflows of AGB stars is the elemental C/O ratio because the atoms of the less abundant element are mostly tied up in the stable CO molecules and do not participate in dust formation (Dorschner & Henning 1996). Therefore, stars with oxygen-rich outflows ($C/O < 1$) produce refractory dust components which are different from the dust condensates of carbon stars ($C/O > 1$). The small transition group of S-type stars is characterized by a C/O ratio very close to 1. As a consequence of nucleosynthesis and dredge-up processes during AGB star evolution, "oxygen-rich" stars with masses smaller than about $4 M_{\odot}$ will evolve into carbon stars and the composition of their molecular outflows will change. Other factors influencing the chemical state of the outflows apart from the C/O ratio can be the formation of molecules by grain surface reactions, the dissociation of some of the CO and N_2 molecules by chromospheric UV radiation, and the influence of shock chemistry. These factors have been suggested to be responsible for the presence of carbon-bearing molecules in the envelopes of individual oxygen-rich late-type stars (see, e.g., Willacy & Millar 1997 and references therein).

In oxygen-rich envelopes, the most abundant elements available for grain formation are O, Fe, Si, Mg, Al, and Ca. Without considering the kinetics of grain formation, we therefore expect the presence of silicates and other oxides, iron and silicon as well as FeSi grains. Spectroscopic evidence for the presence of amorphous silicates comes from the observations of two broad features at 10 and 18 μm in a large number of objects (see the reviews by Dorschner & Henning 1995; Habing 1996). If the feature appears in emission, self-absorption or absorption mainly depends on the optical depth of the envelope (Henning 1983). The Infrared Space Observatory (ISO) led to the clear spectroscopic detection of the presence of crystalline Mg-rich silicates with prominent features between 20 and 45 μm , which vary in strength from object to object (Waters et al. 1996; Cami et al. 1998; Jäger et al. 1998a). Evidence for the presence of Al_2O_3 comes from the observation of a relatively broad feature between 11 and 12 μm and a narrow feature at 13 μm (Onaka et al. 1989; Stencel et al. 1990; Sloan et al. 1996; Begemann et al. 1997 - see also for laboratory data, Justtanont et al. 1998). We should note that the identification of the 13 μm feature remains controversial and that only crystalline $\alpha\text{-Al}_2\text{O}_3$ (corundum) would be a good candidate for the carrier (Begemann et al. 1997). The identification with crystalline $\alpha\text{-Al}_2\text{O}_3$ is supported by the observed correlation of the 13 μm feature with emission around 20 μm (Aringer et al. 1998). Such a correlation is expected because $\alpha\text{-Al}_2\text{O}_3$ has a second band around 20 μm . Based on KAO data, an additional feature at 7.15 μm has been detected, whose carrier remains to be identified (Goebel et al. 1994). In addition, there exists some spectroscopic evidence for the presence of other metal oxides (see Henning et al. 1995a), although the formation of iron oxides seems to be kinetically inhibited in AGB star envelopes (Gail & Sedlmayr 1998a). The formation of oxides other than

aluminium oxides or silicates may, however, occur due to solid-state reactions (Jäger et al. 1998a).

Spectra of carbon-rich stars show two clear dust features peaking at about 11.2 and 30 μm whose strength increases as a function of optical depth (see, e.g., Omont et al. 1995, Speck et al. 1997, Yamamura et al. 1998). The 11.3 μm feature has been attributed to SiC grains (see Mutschke et al. 1998b), whereas the 30 μm feature has been identified with MgS (see Begemann et al. 1994). Features in IRAS LRS spectra at about 8.5 and 11.9 μm have been attributed to hydrogenated amorphous carbon grains (Baron et al. 1987; Goebel et al. 1995). However, such grains produced in an oxygen-free atmosphere do not show a strong band at 8.5 μm . Therefore, the identification of this band remains an open problem. It is striking that the PAHs as the suggested carriers of the ubiquitous “unidentified” IR bands (UIBs) are not observed in carbon-rich AGB stars, although they should be part of the formation route of carbon grains from the acetylene molecule. The lack of evidence for the presence of PAHs in the envelopes of AGB stars may be related to the absence of exciting UV and optical radiation and/or the fact that the PAHs are rapidly consumed during the dust formation process. In the latter case, PAHs would only be produced during later stages of stellar evolution or during processing of grains in the diffuse interstellar medium (see, e.g., Speck & Barlow 1997, Tielens 1997). It is also important to note that absorption features due to aromatic (3.3 μm) or aliphatic (3.4 μm) hydrocarbon grains are not observed. This may be due to the lack of such a component or a band which is weak compared with the continuum (see Schnaiter et al. 1998). The characteristic 3.4 μm absorption feature observed in some post-AGB stars and the diffuse interstellar medium could be the result of a further evolution of aliphatic hydrocarbon grains in a hydrogen-rich environment (Chiar et al. 1998; Schnaiter et al. 1998).

IRAS LRS data already showed that some carbon stars still have silicate features in their spectra, possibly as a fingerprint of their preceding evolutionary phase (Little-Marenin 1986; Willems & de Jong 1988; Chan & Kwok 1991; Kwok & Chan 1993). This detection was later confirmed by ground-based observations (LeVan et al. 1992). Various models have suggested that oxygen-rich material may be present in a long-lived disk which was left over from the “oxygen-rich” phase of the giant (see Jura 1997). Waters et al. (1998) indeed demonstrated the presence of an oxygen-rich circumbinary disk and carbon-rich material in the extended nebula of the Red Rectangle system.

The identification of spectral features with well-defined materials produced in the laboratory requires the derivation of observationally-based mass absorption coefficients or the complex dielectric function of the cosmic materials (see, e.g., Ossenkopf et al. 1992 for silicate data and Begemann et al. 1997 for aluminium oxide data). However, one has to realize that even in the case of optically thin envelopes radiation transfer effects complicate the derivation of such data. A “good” example is the ratio of the strengths of the 10 to 18 μm silicate features, which can only be reliably determined if the dust temperature distribution is given or self-consistently calculated (Henning et al. 1983). In addition, the presence of molecular absorption bands has to be taken into account when deriving the optical properties of CS dust from IR spectroscopy. Additional complicating factors come from the influence of shape/size distribu-

tions and coatings on the spectral appearance of solid-state features, which are especially important for very strong resonances. A textbook example for such a strong resonance is the IR band of crystalline SiC (Bohren & Huffman 1983; Papoular et al. 1998; Mutschke et al. 1998b). Finally, more than one grain component usually contributes to the continuum flux making the derivation of the optical properties for *one* component extremely difficult.

3. Grain formation in AGB star envelopes

The first steps of the kinetic route towards the formation of carbon grains from acetylene have recently been reviewed by Cherchneff (1998). However, specific experiments which relate IR spectra of carbonaceous grains to their formation conditions and internal structures have just started (see, e.g., Schnaiter et al. 1998). Our knowledge of how and which grains form in oxygen-rich environments is mostly guided by thermodynamical arguments or classical nucleation theory (see, e.g., Gail & Sedlmayr 1998a, b). However, non-equilibrium effects (Nuth 1996), kinetic barriers in low-density regions of the outflows (Gail & Sedlmayr 1998a for a discussion of “rusting” of iron in AGB star envelopes), and the details of solid-state reactions (Stencel et al. 1990; Jäger et al. 1998a) must be considered before we can reach a better understanding of the formation of grains in oxygen-rich envelopes. The formation of crystalline silicates by annealing of amorphous structures seems to be possible if the grains are hot enough in the dust formation zone ($T \geq 900$ K) (Gail & Sedlmayr 1998a, b). A discussion of the different possible explanations why the crystalline silicate grains in AGB star envelopes are Mg-rich can be found in Tielens et al. (1998). The formation of hydrated silicates by the reaction with water vapour was generally assumed to be inefficient at the low densities of the outer envelopes. However, the ISO data have to be carefully checked for the signatures of such materials. Grain formation may well start with the nucleation of the most stable clusters which could be TiC clusters (carbon-rich envelopes; Cherchneff et al. 1998) and TiO clusters (oxygen-rich envelopes; Gail & Sedlmayr 1998c) with the subsequent formation and growth of other species on the surfaces of these clusters. In addition, strong evidence exists for the presence of high-density clumps in the AGB star envelopes. In such regions the growth of much larger particles may be possible. Direct evidence for the formation of such large grains comes from the presence of micron-sized SiC stardust grains in primitive meteorites.

4. Laboratory data

The laboratory measurement of optical data for astronomically relevant materials requires three steps: production of the material, detailed characterization of its structure, and the measurement of transmission or reflection data.

A wide variety of techniques has been applied for the preparation of cosmic dust analogues including laser evaporation and pyrolysis (silicon, silicates, carbides, carbonaceous materials), joint evaporation of Mg or FeO and SiO (or O₂ and SiH₄) (silicates), evaporation from an electric arc or resistively heated rods with quenching afterwards (silicates, carbonaceous materials), quenching melts (silicates), sol-gel reactions (silicates), plasma deposition techniques (car-

bon films), and combustion and pyrolysis of organic material (carbonaceous grains). The structure of the material has to be carefully characterized in order to relate optical properties to the structural composition (e.g. silicate band positions as a function of Mg/Fe content, slope of the FIR absorption of carbonaceous grains as function of the sp^2 to sp^3 hybridization ratio). Relevant techniques are SEM/TEM (scanning/transmission electron microscope) analysis and EDX (energy dispersive X-ray diffraction) analysis for silicates (Jäger et al. 1998a) and EELS (electron energy loss spectroscopy) and HRTEM (high-resolution electron microscopy) for carbonaceous materials (Henning & Schnaiter 1999). The last step is the derivation of optical constants or absorption and scattering efficiencies. In the case of measurements on small-particle systems, size, shape, and agglomeration effects have to be carefully taken into account.

The following laboratory sections will concentrate on silicates and carbonaceous materials. Data for other oxides can be found in Begemann et al. (1997) and Henning et al. (1995a). Recent results for SiC grains have been compiled by Papoular et al. (1998) and Mutschke et al. (1998b). These two studies clearly demonstrate that shape effects as well as free charge carriers are much more important than crystal type in determining SiC spectra. A recent experimental study of MgS and FeS can be found in Begemann et al. (1994).

4.1. Silicates

Much of the earlier work on cosmic silicates has been summarized by Dorschner & Henning (1986) and Jäger et al. (1994) and many useful references can be found in these papers.

One of the questions of silicate mineralogy is the problem of non-crystalline silicates having regular silicate-oxygen tetrahedra and consisting of sufficiently homogeneous phases or showing deviations from stoichiometric proportions and considerable compositional variations from point to point. Basic data for the first class of amorphous silicates have been provided by Jäger et al. (1994), Dorschner et al. (1995), and Mutschke et al. (1998a). Nuth & Hecht (1990) introduced the term “chaotic” silicates for the second class of materials. Most experimental data for such solids, also designated as Mg-SiO smokes, have been gathered by B. Donn and J. Nuth (see, e.g., Nuth 1996). The structure of these silicates could be revealed by a combination of SEM/TEM and EDX techniques. However, such data are not always available for the chaotic silicates. Phase separation has been observed in silicate melts during cooling (Jäger et al. 1998a) leading to systems with different well-defined phases, which may finally look like “chaotic” materials on coarser scales.

A special problem of CS silicate dust has always been its “dirtiness”. CS silicate grains have to be much more absorptive at near-infrared (NIR) wavelengths than most of the terrestrial minerals or glasses. In order to explain the observed spectral energy distributions, Jones & Merrill (1977) coined the term “dirty silicates” for materials with high absorption at NIR wavelengths (high value of the imaginary part k of the refractive index; $k=0.1$). Two possibilities exist to produce such dirty silicates: (a) an admixture of finely dispersed impurities of iron or iron oxides to the silicates (Ossenkopf et al. 1992) or (b) the presence of Fe^{2+} cations in the silicates leading to transitions of the 3d electrons in the iron cations (Dorschner et al. 1995). In the latter case a systematic

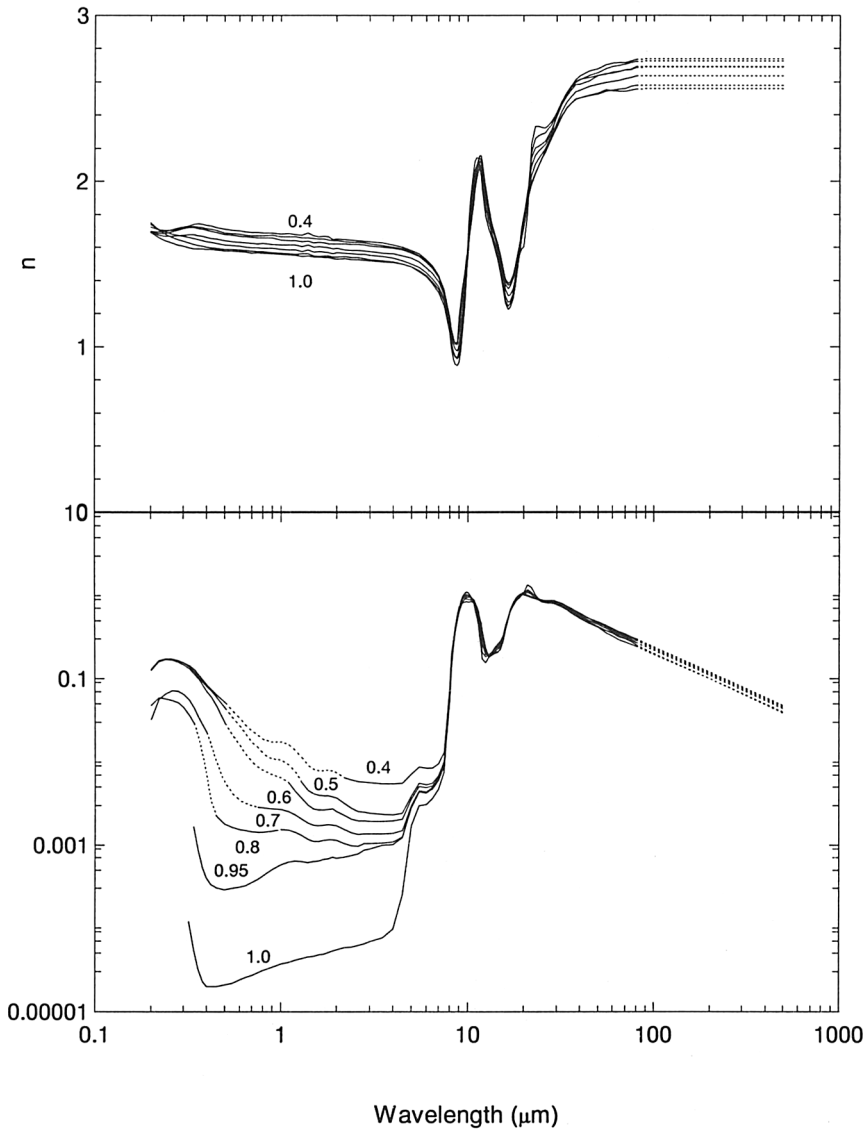


Figure 1. Optical constants n (top) and k (bottom) of pyroxene glasses of the compositions $\text{Mg}_x\text{Fe}_{1-x}\text{SiO}_3$ for $x=0.4, 0.5, 0.6, 0.7, 0.8, 0.95,$ and 1.00 . In both graphs, dotted lines indicate regions of interpolations or extrapolations. After Dorschner et al. 1995.

increase of the NIR absorptivity with iron content has been found in pyroxene glasses (see Fig. 1).

Spectroscopy with ISO clearly revealed the presence of crystalline silicates. Two main groups of astronomically relevant minerals exist: olivines and pyroxenes (see Fig. 2). Olivines can be considered as solid solutions of Mg_2SiO_4 and Fe_2SiO_4 ; the mixture has the sum formula $\text{Mg}_{2x}\text{Fe}_{2-2x}\text{SiO}_4$ with x between 1 and 0. The olivines are neso-silicates (island silicates) because they consist of isolated SiO_4 tetrahedra which are linked by divalent cations. The pyroxenes form a large group of crystalline silicates belonging to the inosilicates (chain silicates). The two SiO_4 tetrahedra form chains, i.e. each tetrahedron shares two of its oxygen atoms with its neighbours. Like in the case of olivines, the symbolic formula $\text{Mg}_x\text{Fe}_{1-x}\text{SiO}_3$ with x between 1 and 0 can be used.

Optical data for these two classes of crystalline silicates with varying Fe/Mg ratios have been provided by Koike et al. (1993) and Jäger et al. (1994, 1998a). An assignment of the bands to the different modes (as far as this is known) is given by Jäger et al. (1998a). IR spectra of pyroxenes are usually more complicated because the polymerization leads to additional bands. The strong olivine bands at 23.5 and 33.5 μm as well as a variety of pyroxene bands could be detected in ISO spectra (Waters et al. 1996). The laboratory data (Jäger et al. 1998) show a clear correlation between Fe/Mg ratio and band position. The bands are shifted towards longer wavelengths with increasing Fe content. The features observed with ISO always point to the presence of the magnesium-rich end members of the silicates. A useful indicator for the presence of Mg-rich olivine is a feature at 69.7 μm which is not present in iron-rich silicates. It is important to note that the observed features are even narrower than the features of the crystalline silicates measured in the laboratory. This may point to the formation of extremely pure crystals in the AGB star envelopes.

4.2. Carbonaceous solids

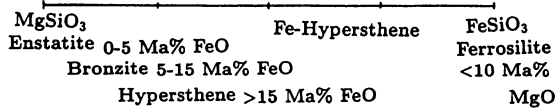
Carbon forms the basis of a rich molecular chemistry in CS envelopes of carbon-rich AGB stars (see, e.g., Oloffson 1997) and leads to the possibility of an enormous structural variety of dust grains (see Henning & Schnaiter 1999, Henning & Salama 1998) because of the unique property of this element to occur in different hybridization states. Carbon atoms in the sp^3 hybridization state are the basis of solids with diamond-like structure. The sp^2 hybridized carbon atoms form planar graphitic structures whose size and distribution depend on the condensation conditions. Apart from the distinct hybridization states, carbon atoms in mixed hybridization states exist which lead to curved structures. In the perfect case these curved structures are closed shells (fullerites). In non-crystalline carbonaceous grains, all kinds of mixtures of, for instance, sp^2 and sp^3 hybridization as well as mixed hybridization states are possible. This leads to carbonaceous materials of different short-, medium-, and long-range order (Papoular et al. 1996; Michel et al. 1998; Jäger et al. 1999; see Henning & Schnaiter 1999 for a review).

Carbonaceous grains dominate the FIR and submm/mm emission of the envelopes of carbon-rich AGB stars. The submm/mm dust continuum radiation is used for the determination of the total mass of the envelopes. This estimate strongly depends on the value of the dust opacity at the wavelength of

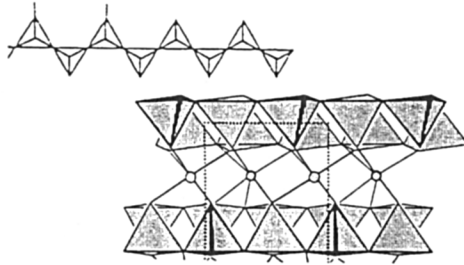
Silicate structure

Pyroxenes: $\text{MgFeSi}_2\text{O}_6$ or
 $\text{Mg}_x\text{Fe}_{1-x}\text{SiO}_3$

Solid solution series
of $\text{MgSiO}_3/\text{FeSiO}_3$:

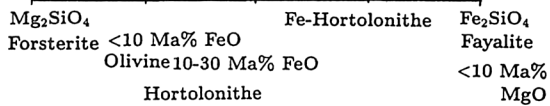


Chain silicates: $[\text{SiO}_3]_{\infty}^{2-}$



Olivines: $\text{Mg}_{2x}\text{Fe}_{2-2x}\text{SiO}_4$

Solid solution series
of $\text{Mg}_2\text{SiO}_4/\text{Fe}_2\text{SiO}_4$:



Neso silicates: $[\text{SiO}_4]^{4-}$

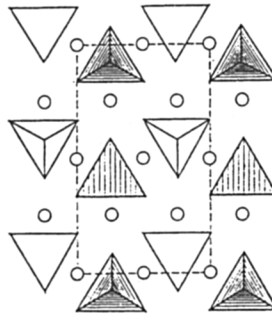


Figure 2. Structure of olivines and pyroxenes

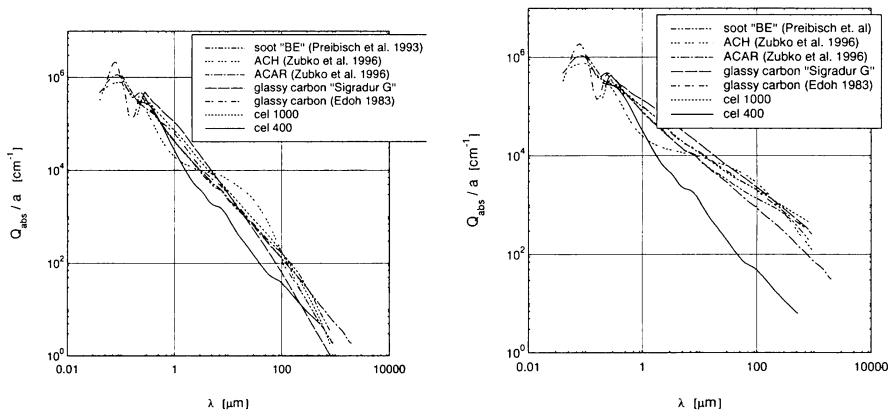


Figure 3. Absorption efficiencies divided by particle radius calculated for spheres in vacuum from the optical data obtained for the materials cel400 (sp² content= 67%) and cel1000 (sp² content= 80%) compared with the results for other soot materials (Preibisch et al. 1993; Zubko et al. 1996) and glassy carbon (Edoh 1983) (left figure). The same calculations for a CDE distribution of particle shapes (right figure). After Jäger et al. (1998b).

the observed flux. Based on the “observed” dust absorption emissivity law for these particles ($Q(\lambda) \propto \lambda^{-\beta}$ with $\beta \simeq 1$; see, e.g., Sopka et al. 1985, Martin & Rogers 1987, Bagnulo et al. 1998 - see also for a discussion of the combined effects of optical properties and geometrical structure of the envelope), it was generally concluded that the carbon grains in these envelopes have a structure characterized by heavily disturbed graphitic zones and larger contents of sp³ hybridized carbon. Therefore, we must understand the relation between the structural properties of carbon grains and the optical behaviour at long wavelengths. Unfortunately, strong IR features due to C-H vibrations, which are of great diagnostic value for the structural characterization, have not been observed in the spectra of AGB stars (see, e.g., Chiar et al. 1998 and Schnaiter et al. 1998 for the discussion of possible reasons). Here, we should stress that the FIR/submm/mm optical properties of carbonaceous grains are not only influenced by their internal structure, but also by the morphology of the grains. It is a well-known fact that the clustering of carbon grains can strongly influence the spectral index β (Rouleau & Martin 1991; Henning et al. 1995b; Stognienko et al. 1995; Henning 1996).

The influence of the internal structure on the FIR behaviour of different carbon materials has been investigated by Rouleau & Martin (1991), Preibisch et al. (1993), Koike et al. (1994, 1995), Menella et al. (1995), Colangeli et al. (1995), and Zubko et al. (1996). These investigations are based on data from transmission spectroscopy. This method cannot exactly separate between structural and morphological effects on the FIR absorption. To overcome this problem, Jäger et al. (1998b) produced well-characterized bulk samples with different sp²/sp³ ratios and obtained optical constants from reflection spectroscopy in the wavelength range between 200 nm and 500 μm . Based on these fundamental data,

an evaluation of the optical behaviour of spherical particles composed of such materials and a continuous distribution of ellipsoids (CDE) was possible (see Fig. 3). The β value for the spheres changed from 1.25 (sp^2 content=67%) to 2.28 (sp^2 content=80%). However, the CDE calculation demonstrated the importance of morphological effects for materials with high sp^2 content. Whereas the β value did not change for the first material, it moved to a value of 0.71 in the second material with the higher sp^2 content.

5. Conclusions

Observations of AGB stars with the ISO spectrometers have revolutionized our understanding of molecular and solid-state chemistry in circumstellar envelopes. However, this progress would not have been possible without high-quality experimental data whose production needs its own effort. Laboratory astrophysics will also contribute to the better understanding of the chemical and physical processes going on in the envelopes.

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