

# Infrared spectra of meteoritic SiC grains

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**Abstract.** We present here the first infrared spectra of meteoritic SiC grains. The mid-infrared transmission spectra of meteoritic SiC grains isolated from the Murchison meteorite were measured in the wavelength range 2.5–16.5  $\mu\text{m}$ , in order to make available the optical properties of presolar SiC grains. These grains are most likely stellar condensates with an origin predominately in carbon stars. Measurements were performed on two different extractions of presolar SiC from the Murchison meteorite. The two samples show very different spectral appearance due to different grain size distributions. The spectral feature of the smaller meteoritic SiC grains is a relatively broad absorption band found between the longitudinal and transverse lattice vibration modes around 11.3  $\mu\text{m}$ , supporting the current interpretation about the presence of SiC grains in carbon stars. In contrast to this, the spectral feature of the large ( $> 5 \mu\text{m}$ ) grains has an extinction minimum around 10  $\mu\text{m}$ . The obtained spectra are compared with commercially available SiC grains and the differences are discussed. This comparison shows that the crystal structure (e.g.,  $\beta$ -SiC versus  $\alpha$ -SiC) of SiC grains plays a minor role on the optical signature of SiC grains compared to e.g. grain size.

**Key words:** infrared: ISM: lines and bands – ISM: lines and bands – ISM: dust, extinction – stars: carbon – stars: atmospheres – stars: abundances

## 1. Introduction

Following the thermodynamical equilibrium calculations of Friedemann (1969a,b) and Gilman (1969) which suggested that silicon carbide (SiC) particles could form in the mass outflow of carbon stars, Hackwell (1972) and Treffers & Cohen (1974) performed infrared spectroscopy of such stars and thereby provided the empirical evidence for the presence of SiC particles in stellar envelopes. A broad infrared emission feature seen in the spectra of many carbon stars, peaking between 11.0 and 11.5  $\mu\text{m}$  is therefore attributed to solid SiC particles and SiC is believed to be a significant constituent of the dust around carbon stars.

Presolar SiC grains have been identified in primitive meteorites (Bernatowicz et al. 1987). Based on isotopic measurements of the major and trace elements in the SiC grains and on models of stellar nucleosynthesis, it is established that a majority of the presolar SiC grains has their origin in the atmospheres of late-type carbon-rich stars (Gallino et al. 1990, 1994; Hoppe et al. 1994). For recent reviews see, e.g., Anders & Zinner (1993), Ott (1993) and Hoppe & Ott (1997). The grain sizes of presolar SiC from the Murchison meteorite have been found by Amari et al. (1994) to vary from less than 0.05 to 20  $\mu\text{m}$  in equivalent spherical diameter, with about 95% (by mass) of the grains being between 0.3 and 3  $\mu\text{m}$ . This distribution is coarser than for presolar SiC found in other meteorites (Russel et al. 1993, 1997; Huss & Lewis 1995; Gao et al. 1996). Therefore, Russel et al. (1997) have speculated that the finer grained SiC was lost through size sorting in the solar nebula prior to accretion of Murchison. In the carbonaceous chondrites only approximately 0.004% of the silicon is in the form of SiC (the remainder being in the form of silicates).

Silicon carbide occurs in a large variety of crystal types. The basic units are Si-C bilayers with a three-fold symmetry axis, in which the Si and C atoms are closely packed. It is the special stacking of these layers that determines the occurrence of the polytypes. The second bilayer is shifted in the  $[\bar{1}100]$ -direction by  $1/\sqrt{3}$  of the Si-Si or C-C atomic distance in the layer. If a third and a fourth layer is stacked in an identical way, then the atoms in the fourth layer lie exactly above the ones in the first layer. Further repetition of this sequence results in a cubic crystal structure called  $\beta$ -SiC. If at least one bilayer is shifted in the opposite direction the resulting structure is hexagonal or rhombohedral (Mutschke et al. 1999). All the polytypes resulting from non-cubic stacking sequences are summarized in the term  $\alpha$ -SiC.

Virag et al. (1992) have investigated the crystal structure of the large presolar SiC grains extracted by Amari et al. (1994) (LS and LU series). The authors investigated forty-one large (from  $1 \times 1.5 \mu\text{m}$  to  $23 \times 23 \mu\text{m}$ ) grains from the Murchison meteorite by Raman spectroscopy. Thirty-two of these grains were found to have a cubic crystallographic structure ( $\beta$ -SiC), the remaining grains showed a non-cubic structure (hexagonal or rhombohedral;  $\alpha$ -SiC). However, the  $\alpha$ -SiC grains were also characterized by a normal isotopic composition, indicating that

they might not be of presolar origin. Recently Daulton et al. (1998) investigated one of the finer grained samples extracted by Amari et al. (1994) (KJB grain size 0.3–0.7  $\mu\text{m}$ ) and found that for these smaller grains there seems to be even amounts of  $\alpha$ - and  $\beta$ -SiC. This indicates that while the larger presolar grains seem to be dominated by the  $\beta$ -SiC type the smaller grains are a mix of the  $\alpha$ - and  $\beta$ -SiC type, but the presolar nature of the small  $\alpha$ -SiC grains still needs to be confirmed. Whether presolar SiC grains will turn out to be of predominately one or the other crystal type can place constraints on the formation parameters of the grains. Therefore, it has been attempted by several groups to derive the crystal type of circumstellar SiC grains from their observed IR emission spectra (e.g. Blanco et al. 1994, 1998; Groenewegen 1995; Speck et al. 1997). However, it is argued by Papoular et al. (1998) that the IR band profiles might not be sensitive to the crystal type but to other structural and morphological grain properties. To decide these questions, more laboratory studies are needed (Mutschke et al. 1999).

We have previously published the absorption coefficients of presolar diamonds (Mutschke et al. 1995; Andersen et al. 1998) and along this line of providing the spectral feature of “real star dust”, we have now measured the spectral appearance of presolar SiC grains. These data are necessary in order to determine if the spectral emission (and in a few cases absorption (Jones et al. 1978; Speck et al. 1997)) feature found in circumstellar envelopes of carbon stars, is consistent with the meteoritic grains having originated in such stars. Agreement between the isotopic composition of the presolar SiC grains and those predicted in AGB star nucleosynthetic models, give strong belief that these meteoritic grains originated in carbon stars. It is our hope that comparison between the optical properties of presolar SiC and the appearance of the dust features in carbon star spectra will be able to impose further constraints. In this paper, we present results from measurements of the spectral properties of meteoritic SiC in the wavelength range between 2.5 and 16.5  $\mu\text{m}$ .

## 2. The extraction procedure

Two fractions of 26.8 g (sample I) and 10.0 g (sample II) from an originally large (100 g) piece of Murchison (a CM2 chondrite) was used for the extraction of meteoritic SiC grains. The originally large piece was crushed with a steel mortar, to obtain smaller fractions.

To obtain a large and clean sample of the SiC grains for the spectroscopic measurement, it is necessary to isolate the presolar grains from the rest of the meteoritic material. A physical, non-destructive separation does not work well because the grains are tiny and cling to the much larger amounts of fine-grained clay minerals and kerogen (macromolecular organic matter). Therefore, a destructive chemical separation in which undesirable minerals are dissolved by appropriate reagents is necessary. The chemical separation procedures used in this study were variants of those developed by Tang & Anders (1988) and Amari et al. (1994), which mainly consist of a set of progressively more corrosive “selective solvents” to remove the mineral phases one by one, but also includes density separations.

Extracting presolar SiC from meteorites is much more complicated than extracting the presolar nano-diamonds, because the SiC (6 ppm) is much rarer than the nano-diamonds (500 ppm) (Amari et al. 1994), and because the diamonds can be density separated out of the meteoritic sample without having to remove all mineral phases first. It is therefore much harder to obtain a clean sample of presolar SiC grains than of presolar diamonds. There will in most cases always be smaller amounts of corundum, hibonite, spinel and chromite grains present, which are very hard to get rid of without also losing the SiC grains.

The extraction treatment was primarily designed to result in a fraction of very clean presolar nano-diamonds (Braatz in prep.). The individual extraction steps for the two samples were almost identical, except that for the first two steps sample I was treated in a sealed Teflon bomb at 180°C while sample II had a more gentle treatment without the Teflon bomb and at 80°C. The extraction steps were the following: 1) solution of concentrated HCl (32%) to dissolve metals and sulfides; 2) alternating treatment with concentrated HF (48%) and concentrated HCl to dissolve silicates; 3) extraction of precipitated sulfur with CS<sub>2</sub>; 4) oxidation with 0.5 N Na<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> + 2 N H<sub>2</sub>SO<sub>4</sub> at 80°C; 5) extraction of the nano-diamonds as a colloid with a solution of bidistilled water and NH<sub>3</sub> (pH 9–10). This step results in a precipitate at the bottom of the container containing mainly residual spinels and SiC. The nano-diamonds were used for other studies (Braatz in prep.). The spinel-SiC residue was further treated by; 6) boiling in 70% HClO<sub>4</sub> (200°C) to remove residual organic material and graphite; 7) boiling in H<sub>2</sub>SO<sub>4</sub> (180 °C) to remove the spinels (spinel MgAl<sub>2</sub>O<sub>4</sub> and chromite FeCr<sub>2</sub>O<sub>4</sub>) and 8) another treatment with HF/HCl (60°C) to remove remaining silicon bearing grains other than SiC.

Most of the treatments were carried out in Teflon containers. Between all steps intensive washing was carried out, with diluted HCl (pH < 2) and/or bidistilled water, which is important in order to remove possible coatings that impede further reactions (Amari et al. 1994). Each time the supernatant liquid was pipetted into separate discard tubes for the two samples and all solids which settled down in the discard tubes were returned to the main sample.

## 3. Spectroscopy

### 3.1. Measurement procedures

Several groups have carried out experimental work on the spectroscopic properties of commercially available and laboratory produced SiC particles. Stephens (1980) has studied laser-produced  $\beta$ -SiC condensates, Friedemann et al. (1981) measured spectra of commercially available  $\alpha$ -SiC, Borghesi et al. (1985) studied commercially produced  $\alpha$ -SiC and  $\beta$ -SiC, Kaito et al. (1995) have studied  $\alpha$ -SiC and  $\beta$ -SiC produced by simultaneous evaporation of silicon and carbon and Papoular et al. (1998) investigated two samples of  $\beta$ -SiC powders, one commercially available and one produced by laser pyrolysis.

All the groups have done their measurements by embedding the sub-micrometer particles in a solid matrix either of KBr or CsI. In the KBr/CsI pellet technique, small quantities of the

sample are mixed thoroughly with powdered KBr/CsI. Due to the softness of the matrix material and its bulk transparency in the mid-IR, the material can be pressed into a clear pellet.

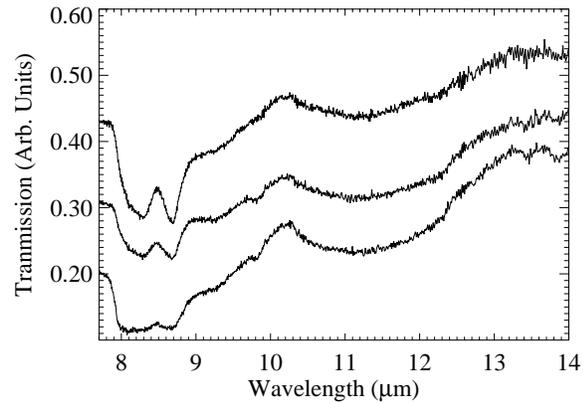
According to scattering calculations, embedding the sample in a matrix will influence the wavelength at which the frequency-dependent extinction falls as well as the intensity, in a way which depends on the sample considered and the matrix in which it is included (Bohren & Huffman 1983; Papoular et al. 1998; Mutschke et al. 1999).

Friedemann et al. (1981) and Borghesi et al. (1985) tried to correct the influence of the matrix by blue-shifting the whole feature by an amount of  $\delta\lambda = -0.4 \mu\text{m}$  and  $\delta\lambda = -0.1 \mu\text{m}$ , respectively. They also corrected the intensity by a factor of 0.7 and 0.9, respectively. These procedures have been argued by Papoular et al. (1998) to be incorrect, since independent of the matrix material, absorption should not fall outside the longitudinal and transverse optical phonon frequencies. Instead Papoular et al. (1998) proposed a new method for computing the expected spectrum for the particles in vacuum, which works if the dielectric function of the grains can be described by a single Lorentzian oscillator. This certainly is the case for SiC (Mutschke et al. 1999).

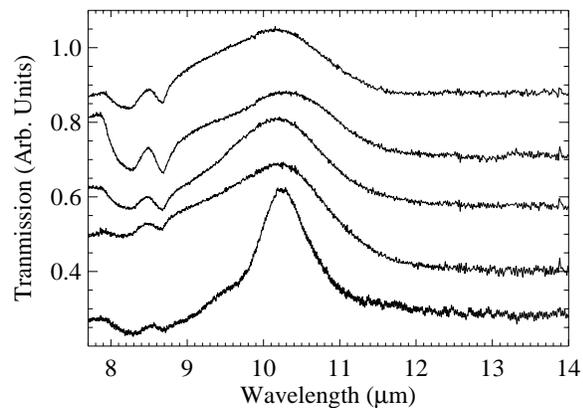
Our transmission measurements were performed by placing the presolar SiC grains on a polished Si substrate. This means that the grains were mainly but not fully surrounded by air, resulting in much less matrix effects than if the sample had been embedded in a solid medium. Samples tend to cluster both in the KBr pellets and on a substrate and may do this with different cluster morphologies. At the moment theory is not able to determine what the optical effect of clustering is for SiC grains. We have not been able to find any systematic change of band profiles as a result of clustering. We have also not found systematic changes of band profiles related to whether we used a Si, NaCl or a KBr substrate (refractive index: KBr and NaCl ( $n=1.5$ ), Si ( $n=3.4$ )). The samples were mounted on the Si substrates by dispersing them in a droplet of chloroform. All the spectroscopic measurements were made with an infrared microscope attached to a Bruker 113v Fourier Transform Infrared Spectrometer. The detector is a liquid-nitrogen-cooled mercury cadmium telluride detector with a spectral range of  $7000\text{--}600 \text{ cm}^{-1}$ . The sampling diameter of the microscope can be as small as  $30 \mu\text{m}$ . Spectra were obtained on different grain clusters of the samples which were 10 to  $80 \mu\text{m}$  in size. The microscope aperture used for the measurements was always  $80 \mu\text{m}$  since this gave a sufficient signal-to-noise ratio with a reasonable number of scans (64). The measurements were performed with a resolution of  $1 \text{ cm}^{-1}$ . The reference spectra were obtained on a blank part of the substrate.

### 3.2. Results

One of the drawbacks with using the infrared microscope is that a reliable mass estimate of the fraction responsible for the spectral feature cannot be obtained. With the microscope one measures different parts of the sample and depending on how good the sample was mounted the mass can be more or less



**Fig. 1.** Infrared spectra of meteoritic SiC grains (sample I) from the Murchison meteorite obtained with the IR microscope at an aperture of  $80 \mu\text{m}$ . The SiC feature is located at about  $11.3 \mu\text{m}$ . The features around  $8.2$  and  $8.6 \mu\text{m}$  are due to Teflon (see text).

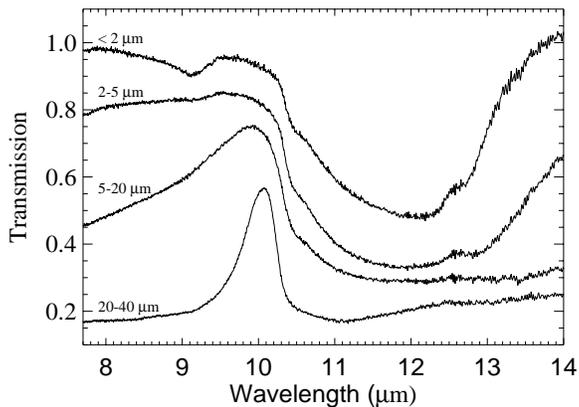


**Fig. 2.** Infrared spectra of meteoritic SiC grains (sample II) from the Murchison meteorite obtained with the IR microscope at an aperture of  $80 \mu\text{m}$ . The appearance of the feature is influenced by the Christiansen effect (see text).

evenly distributed on the substrate. Different densities will result in different depths of the features. The results shown in Fig. 1 for sample I and in Fig. 2 for sample II are the raw data ( $y$  is shifted for better comparison). There is a remarkable difference between the spectral appearance of the SiC grains in the two samples, despite the fact that they were prepared by using almost identical extraction procedures and came from the same larger piece of meteorite. This difference can be explained by different grain sizes (see later).

Fig. 1 (sample I) shows the expected broad absorption feature of small SiC grains between the positions of the longitudinal and transverse lattice vibration modes at  $\omega_L \approx 10.3 \mu\text{m}$  and  $\omega_T \approx 12.6 \mu\text{m}$  (Mutschke et al. 1999). The center of the band is located at about  $11.3 \mu\text{m}$ . The peaks around  $8.2$  and  $8.6 \mu\text{m}$  are due to  $-\text{CF}$  and  $-\text{CF}_2$  groups originating from the treatment in Teflon containers with hot concentrated sulphuric acid ( $\text{H}_2\text{SO}_4$ ). The sulphuric acid did not only dissolve spinel grains but also attacked the walls of the container.

As an attempt to try to get rid of the Teflon-related contaminate, we considered a density separation of the Teflon



**Fig. 3.** Spectral appearance of commercially available  $\alpha$ -SiC (Duisburg) sedimented to obtain different grain size fractions. The variation of the spectral appearance as a result of different grain size is apparent. As the grain size increases the influence of the Christiansen effect becomes important (see text).

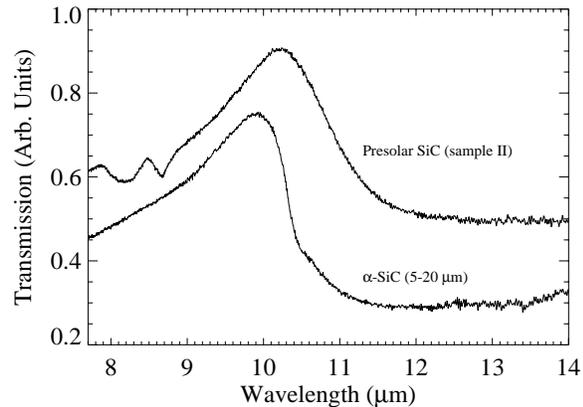
and the SiC grains by use of e.g. sodium-polywolframate ( $3\text{Na}_2\text{WO}_4 \cdot 9\text{WO}_3 \cdot \text{H}_2\text{O}$ ) (Amari et al. 1994). However, all possible solvents, that we could think of, tended to have IR features in exactly the same infrared spectral region as the SiC feature we were interested in. Therefore, we did not follow this approach.

In Fig. 2 (sample II) there is apparently no similar SiC band. Instead one observes a maximum of the transmission spectrum more or less at the place of the longitudinal lattice vibration mode. At longer wavelengths the spectrum is flat. In the next section we will explain that this does not mean the absence of SiC grains.

### 3.3. Size effects

Due to collective processes very small solid particles (small compared to the wavelength) exhibit strong resonances in absorption in the spectral regions where the real part of the dielectric function ( $\epsilon_1$ ) is negative. The precise positions of these resonances, called surface modes, depend on the particle shape, size, and on the nature and amount of coatings or matrixes surrounding the grain (Bohren & Huffman 1983). There are two distinct energy ranges in which resonances occur. One is in the infrared in the region of strong lattice bands between the transverse optical phonon frequency ( $\omega_T$ ) and the longitudinal optical phonon frequency ( $\omega_L$ ). The other is in the ultraviolet and is due to the transitions of bound electrons.

For the surface modes of very small SiC particles in the infrared, Gilra (1973) performed Mie calculations for different shapes and found that for a thin disc the resonances are at  $10.3 \mu\text{m}$  ( $\omega_L$ ) and  $12.6 \mu\text{m}$  ( $\omega_T$ ). As the oblateness decreases, the resonances move towards each other and finally for the spherical case there is only one resonance at  $10.73 \mu\text{m}$  ( $\epsilon_1 = -2$ ). As the particles become prolate, the resonances move away from each other and finally for the case of a needle they are at  $12.6 \mu\text{m}$  ( $\omega_T$ ) and  $10.55 \mu\text{m}$  ( $\epsilon_1 = -1$ ). Gilra (1973) concludes that if the particles are highly irregular there will be a



**Fig. 4.** Spectra of meteoritic SiC grains (sample II) and commercially available  $\alpha$ -SiC (Duisburg) with grain sizes  $5\text{--}20 \mu\text{m}$ .

broad feature between about  $10.3 \mu\text{m}$  and  $12.6 \mu\text{m}$ , which agrees very well with the later findings of Bohren & Huffman (1983) resulting in the continuous distribution of ellipsoids (CDE) approximation.

If the grain size is larger than or comparable to the wavelength, scattering becomes more important and diminishes the transmitted light also at frequencies outside the absorption band. At a special (larger) frequency outside but close to the absorption band,  $n \simeq 1$  and  $k \simeq 0$ . Since these values are close to those of the surrounding air (or free space) then at this spectral point the particles are nearly invisible. In other words, scattering is greatly reduced and a maximum in the transmission spectrum is observed. This is known as the Christiansen effect (Bohren & Huffman 1983).

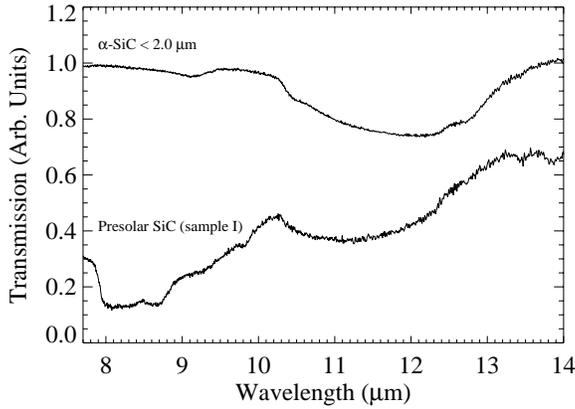
In order to investigate these size effects we did infrared microscope measurements on a commercially available  $\alpha$ -SiC grain sample (Duisburg) with large grain size of up to  $40 \mu\text{m}$ . This sample was sedimented in acetone and based on settling rates calculated from Stokes law

$$t = 18 \frac{h \cdot \mu_{dyn}}{g \cdot d^2 \cdot (\rho_{grain} - \rho_{liquid})} \quad (1)$$

different size fractions were obtained. Here  $t$  is the settling time,  $\mu_{dyn}$  is the dynamic viscosity,  $h$  the settling height,  $g$  the gravitational constant,  $d$  the grain diameter,  $\rho_{grain}$  the density of the grains and  $\rho_{liquid}$  the density of the liquid. The size fractions that were obtained were  $40\text{--}20 \mu\text{m}$ ,  $20\text{--}5 \mu\text{m}$ ,  $5\text{--}2 \mu\text{m}$  and  $< 2 \mu\text{m}$ . These samples have been mounted on Si substrates in the same way as for the meteoritic samples.

In Fig. 3 it is shown that the spectral appearance of SiC grains strongly depends on the grain size and that the Christiansen effect dominates for the larger grains. This means that the absorption feature between  $\omega_T$  and  $\omega_L$  is transformed into a flat spectrum with a transmission maximum at about  $\omega_L$  (see above).

Fig. 4 shows sample II of the meteoritic SiC compared to the  $\alpha$ -SiC (Duisburg) sample with grain sizes  $5\text{--}20 \mu\text{m}$ . Fig. 5 shows sample I of the meteoritic SiC grains compared to the same material but with grain sizes  $< 2 \mu\text{m}$ . These comparisons show that the meteoritic SiC grain spectra look like the profiles



**Fig. 5.** Spectra of meteoritic SiC grains (sample I) and commercially available  $\alpha$ -SiC (Duisburg) with grain sizes less than  $2\ \mu\text{m}$ .

expected from SiC grains of different sizes. It may seem odd that an almost identical extraction procedure on two parts of originally one piece of the Murchison meteorite should result in two samples with different size distributions. However, already the results of Amari et al. (1994) indicate an unusual grain size distribution of extracted Murchison SiC when compared with (less processed) SiC residues from other meteorites (see discussion in Russel et al. 1997). In addition, we cannot exclude that we lost (presumably mostly the finer) grains during our own extensive extraction procedure. A hint that this may have happened comes from the noble gas analysis of one of the diamond fractions, in which we found small, but easily detectable amounts of Ne-E(H), indicating the presence of small amounts of presolar SiC in the nominal diamond fractions.

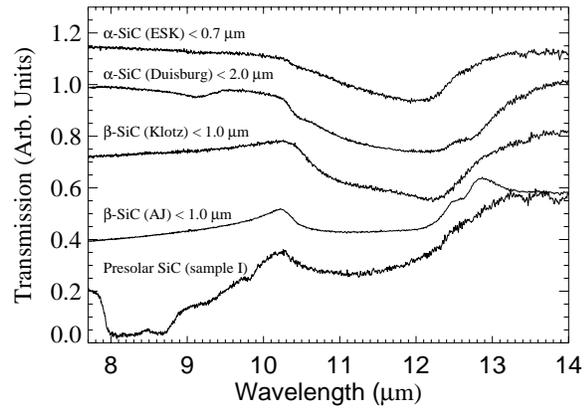
It was not possible to obtain a reliable grain size estimate of the samples by the use of a scanning electron microscope (SEM) due to clustering of the grains. Sample I looks like small grains ( $< 1\ \mu\text{m}$ ) mixed with Teflon and some of the more resisting meteoritic grains such as spinel, chromite, hibonite and corundum, while sample II looks like larger grains in a cleaner environment.

The different Christiansen frequencies observed for the meteoritic and the commercial SiC grains (Fig. 4) hint to different optical constants for the two samples. However, so far we have not been able to correlate this to a certain polytype of the meteoritic SiC grains.

The measured spectra of sample II (Fig. 4) can therefore be understood as being a spectrum of large meteoritic SiC grains, where instead of an extinction maximum around  $11.3\ \mu\text{m}$  we see an minimum around  $10\ \mu\text{m}$ . The measured spectra of sample I (Fig. 5) corresponds to a spectrum of mainly small ( $< 2\ \mu\text{m}$ ) meteoritic grains.

### 3.4. Comparison with $\alpha$ - and $\beta$ -SiC samples

In the last years, there have been quite a number of papers addressing the structural differences of SiC particles of different polytype as the main important factor influencing the band profile and the peak position of the  $11.3\ \mu\text{m}$  feature observed in



**Fig. 6.** Presolar SiC plotted with different commercially available  $\alpha$ - and  $\beta$ -SiC samples with the grain sizes indicated. More information about the samples is given in Mutschke et al. (1999).

carbon stars (e.g. Blanco et al. 1994, 1998; Groenewegen 1995; Speck et al. 1997). In another paper (Mutschke et al. 1999) we study these spectral differences due to the polytype in detail. Here, we present infrared microscope measurements on four commercially available SiC powders – two  $\alpha$ -SiC and two  $\beta$ -SiC – and compare them to the spectrum of sample I (Fig. 6).

The five spectra presented in Fig. 6 all show the SiC infrared band of small grains between  $\omega_L \approx 10.3\ \mu\text{m}$  and  $\omega_T \approx 12.6\ \mu\text{m}$ . Obviously, there are significant differences in band shape and peak position between the spectra. However, these differences are not related to the polytypes. The meteoritic spectrum resembles most the  $\beta$ -SiC (AJ) band profile but this definitely cannot be taken as an indication of the polytype of the meteoritic grains as one can see by comparing with the spectrum of  $\beta$ -SiC (Klotz) in Fig. 6.

This is consistent with the findings of Papoular et al. (1998) and Mutschke et al. (1999) that the band profile and consequently the peak wavelength of the SiC infrared band depends on the distribution of shapes and grain sizes rather than crystal type. Therefore using the IR spectral feature of different polytypes to determine whether one of the other crystal type of SiC dominates in circumstellar outflow, cannot be recommended.

The fact that commercially available SiC samples vary so much in spectral appearance strengthens the importance of studying the spectral feature of presolar SiC. However, so far it seems that from the band profiles we will learn rather about grain shape and size than about the polytype of extra-solar grains. In any case further intensive laboratory studies are needed. A step towards a better understanding will be presented in Mutschke et al. (1999).

## 4. Summary and conclusions

The discovery of meteoritic dust grains with an origin outside the solar system has opened the possibility of studying presolar material directly in the laboratory. A large fraction of this material is likely to be dust from the envelopes of asymptotic giant branch (AGB) stars. We have previously measured the absorption coefficients of presolar diamonds (Mutschke et al. 1995;

Andersen et al. 1998). We here report the results of mid-IR measurements of meteoritic SiC grains.

Measurements were performed on two different extractions of presolar SiC from the Murchison meteorite. The two samples show very different spectral appearances which we interpret as being due to different grain size distributions in the two extractions. The spectral feature of the smaller meteoritic SiC grains is at  $11.3\ \mu\text{m}$ , whereas the large ( $> 5\ \mu\text{m}$ ) SiC grains have no extinction maximum at  $11.3\ \mu\text{m}$ , but instead are characterized by an extinction minimum around  $10\ \mu\text{m}$ . It is a common interpretation that the  $11.3\ \mu\text{m}$  band observed in carbon stars is due to SiC dust. It is also a common interpretation (based on comparison of isotopic ratios in the meteoritic SiC and nucleosynthesis models of AGB stars) that the majority of the presolar SiC grains come from carbon stars. If both of these common interpretations are correct, we conclude from our measurements that the  $11.3\ \mu\text{m}$  feature in carbon stars can be understood as being caused by the smaller end of the size distribution of SiC of the type identified in primitive meteorites, and the larger grains must correspond to a grain distribution not yet identified in carbon stars.

In the observational data by Speck et al. (1997) the (interpreted) SiC feature in carbon star spectra peak around  $11.3\ \mu\text{m}$  for about 40% of cases (13 out of the sample of 30 stars) with nearly symmetric profiles and a FWHM around  $1.8\ \mu\text{m}$ . These features, which are relatively broad just as the feature of the small meteoritic SiC (sample 1), may be interpreted as an indication that the circumstellar SiC is of this small grain size. If the grain size distribution evolves towards larger grains, for example during later stages of the carbon star evolution, it would result in a weakening (and possible disappearance) of the  $11.3\ \mu\text{m}$  feature, which could explain the remaining 60% of stellar spectra. However, there are several strong molecular features in the  $10\text{--}14\ \mu\text{m}$  area in carbon stars (Hron et al. 1998), and a unique interpretation of the observational data still awaits a self-consistent simulation taking both the molecular absorption and the dust emission into account for a wide range of types of carbon stars.

The fact that large ( $> 5\ \mu\text{m}$ ) SiC grains have a different spectral appearance than smaller ( $< 2\ \mu\text{m}$ ) SiC grains, will make large SiC grains difficult to observe in the interstellar medium because of the presence of silicate-related absorption around  $10\ \mu\text{m}$ . If the majority of the cosmic SiC grains have the size distribution found by Amari et al. (1994), this will probably not significantly change the abundance limit of less than a few percent SiC compared to silicates in the ISM (Whittet et al. 1990),

The results of spectral measurements on commercially available SiC grain samples of different polytypes and the meteoritic SiC grain samples show that the variations among polytypes of SiC grains are smaller than the variations due to different grain size. It is therefore not possible to distinguish, by IR spectroscopy, between  $\alpha$ - and  $\beta$ -SiC of dusty material as also discussed by Papoular et al. (1998) and Mutschke et al. (1999).

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## References

- Amari S., Lewis R.S., Anders E., 1994, *Geochim. Cosmochim. Acta* 58, 459
- Anders E., Zinner E., 1993, *Meteoritics* 28, 490
- Andersen A.C., Jørgensen U.G., Nicolaisen F.M., Sørensen P.G., Glejbjøl K., 1998, *A&A* 330, 1080
- Bernatowicz T., Fraundorf G., Tang M., et al., 1987, *Nat* 330, 728
- Blanco A., Borghesi A., Fonti S., Orofino V., 1994, *A&A* 283, 561
- Blanco A., Borghesi A., Fonti S., Orofino V., 1998, *A&A* 330, 505
- Bohren C.F., Huffmann D.R., 1983, *Absorption and Scattering of Light by Small Particles*. John Wiley & Sons, New York
- Borghesi A., Bussoletti E., Colangeli L., De Blase C., 1985, *A&A* 153, 1
- Daulton T.L., Lewis R.S., Amari S., 1998, *Meteoritics & Planetary Science* 33, A37
- Friedemann C., 1969a, *Physica* 41, 139
- Friedemann C., 1969b, *Astron. Nach.* 291, 177
- Friedemann C., Gürtler J., Schmidt R., Dorschner J., 1981, *Ap&SS* 79, 405
- Gallino R., Busso M., Picchio G., Raiteri C.M., 1990, *Nat* 348, 298
- Gallino R., Raiteri C.M., Busso M., Matteucci F., 1994, *ApJ* 430, 858
- Gao X., Amari S., Messenger S., et al., 1996, *Meteoritics & Plan. Sci. Supp.* 31, A48
- Gilman R.C., 1969, *ApJ* 155, L185
- Gilra D.P., 1973, *Dust Particles and Molecules in the Extended Atmosphere of Carbon Stars*. In: Greenberg J.M., Van de Hulst H.C. (eds.) *Interstellar Dust and Related Topics*. IAU Symposium 52, Reidel, Dordrecht, p. 517
- Groenewegen M.A.T., 1995, *A&A* 293, 463
- Hackwell J.A., 1972, *A&A* 21, 239
- Hoppe P., Ott U., 1997, *Mainstream Silicon Carbide Grains from Meteorites*. In: Bernatowicz T., Zinner E. (eds.) *Astrophysical Implications of the Laboratory Study of Presolar Materials*. AIP 402, New York, p. 27
- Hoppe P., Amari S., Zinner E., Ireland T., Lewis R.S., 1994, *ApJ* 430, 870
- Hron J., Loidl R., Höfner S., et al., 1998, *A&A* 335, L69
- Huss G., Lewis R.S., 1995, *Geochim. Cosmochim. Acta* 59, 115
- Jones B., Merrill K.M., Puetter R.C., Willner S.P., 1978, *AJ* 83, 1437
- Kaito C., Nakamura H., Sakamoto T., et al., 1995, *Planetary & Space Science* 43, 1271
- Mutschke H., Dorschner J., Henning Th., Jäger C., Ott U., 1995, *ApJ* 454, L157
- Mutschke H., Andersen A.C., Clément D., Henning Th., Peiter G., 1999, submitted to *A&A*
- Ott U., 1993, *Nat* 364, 25
- Papoular R., Cauchetier M., Begin S., LeCaer G., 1998, *A&A* 329, 1035
- Russel S.S., Alexander C.M.O'D., Ott U., et al., 1993, *Meteoritics* 28, 425
- Russel S.S., Ott U., Alexander C.M.O'D., et al., 1997, *Meteoritics & Plan. Sci.* 32, 719
- Speck A.K., Barlow M.J., Skinner C.J., 1997, *MNRAS* 288, 431
- Stephens J.R., 1980, *ApJ* 237, 450
- Tang M., Anders E., 1988, *Geochim. Cosmochim. Acta* 52, 1235
- Treffers R., Cohen M., 1974, *ApJ* 188, 545
- Virag A., Wopenka B., Amari S., et al., 1992, *Geochim. Cosmochim. Acta* 56, 1715
- Whittet D.C.B., Duley W.W., Martin P.G., 1990, *MNRAS* 244, 427