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Research Note

Optical properties of α silicon carbide

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Summary. Laboratory extinction spectra of grains of α silicon carbide are available which compare well with the astronomical observations. Unfortunately, these measurements are not sufficient to fully interpret the 11.5 μm feature profiles since radiative transfer models require the knowledge of the complex refractive index of the circumstellar material over the whole electromagnetic spectrum. In order to fill in this gap, we present in this work a complete dielectric function for this material computed from a Kramers-Kronig analysis of the above mentioned extinction spectra and other measurements of the index of refraction. These synthetic optical properties are shown to be satisfactorily consistent with the existing laboratory measurements and confirm the assignment of the feature exhibited by C-stars spectra to α–SiC.

Key words: infrared radiation – interstellar medium: dust – stars: carbon – stars: late-type

1. Introduction

The computations of Friedemann (1969) and Gilman (1969) have shown that silicon carbide is a valuable candidate as a component of dust around carbon stars. This theoretical prediction was strongly supported by the discovery by Hackwell (1972) of an emission feature at 11.5 μm in the spectra of some C-stars, the profile of which was investigated by Treffers and Cohen (1974) and Merrill and Stein (1976). Its assignment to solid SiC was confirmed by Goebel et al. (1980) who succeeded in fitting the 11.5 μm band of Y Canis Venaticorium, using laboratory measurements of Dorschner et al. (1977). However, the total number of available spectra in the 10 μm region remained rather small and it is only with the publication of the IRAS results (542 spectra of C-stars exhibiting the 11.5 μm feature in the class 4n of the LRS catalog, see Olnon, 1985) that it became possible to have an overview of the general behaviour of this band. Modelling these observations can lead to important information on the physical parameters characterizing these objects (optical depth and inner radius of the shell, size and temperature of grains) but requires the knowledge of the scattering and absorption properties of the different populations of grains over the whole electromagnetic spectrum to correctly describe the radiative transfer in the shell. Furthermore, a statistical study of the LRS spectra (Baron et al., 1987) pointed out strong correlations between the strength and shape of the SiC band and the strength of secondary features and/or the slope of the underlying continuum attributed to thermal emission of graphite and/or amorphous carbon grains. A complete interpretation of such correlations would also require the knowledge of the optical properties of the above mentioned materials. Unfortunately, while refractive indices are given for graphite by Draine and Lee (1984) and by Hagemann et al. (1974, 1975) for a-C, there are no satisfactory optical properties of SiC available in literature, although numerous laboratory works have been devoted to this material, due to its technological importance. This work is an attempt to derive such a comprehensive dielectric function from a Kramers–Kronig analysis of a compilation of partial data from different authors (presented in Sect. 2). The procedure used to determine these optical properties is described in Sect. 3 and the results discussed in Sect. 4.

2. The available laboratory measurements

Silicon carbide exists in two cristallographic forms: hexagonal/rhomboedric (α-Sic), which exhibits a strong absorption peak around 11.3 μm, and cubic (β-SiC), the absorption profile of which is more variable and can present a double peak (see Stephens, 1980). Although Cohen (1984) finds that some particular spectra can be interpreted in terms of these two profiles, a recent analysis of the IRAS C-stars spectra does not support this point and indicates that α-SiC is the best candidate to reproduce the astronomical observations in the 10 μm region (Baron et al., 1987). We therefore concentrate on it in this work. The numerous laboratory data concerning this material are mainly of two sorts: explicit optical properties in a given range of wavelength, which are generally obtained from an analysis of reflection and transmission measurements of thin films of material; and extinction spectra of powders, in vacuum or embedded in some convenient matrix.

i) Laboratory measurements of the complex refractive index of α-SiC are available in the visible and ultraviolet (Phillip and Taft, 1960) and, for its real part only, in the visible and near-infrared (Thibault, 1944; Choyke and Patrick, 1968). Up to 40 μm, measurements were also performed by Pikhtin et al. (1977) who obtained quite similar values for both $E \parallel c$ and $E \perp c$. However, in this latter case, the determination of $n(\lambda)$ is not complete in the vicinity of the strong band at ~11.5 μm. In this region, indices are given by Spitzer et al. (1959), but with values so high (n and $k \sim 17$ at the absorption peak) that it seems difficult to build a consistent dielectric function from these results and that of the previous authors.

ii) In the mid-infrared, after the first laboratory works of Dorschner et al. (1977), mutually consistent absorption spectra were measured by Friedemann et al. (1981) and Borghesi et al. (1983) between 2.5 and 40 μm. These results were extended up to 300 μm by Borghesi et al. (1986) and the behaviour of the 11.5 μm band thoroughly discussed in Borghesi et al. (1985). In this last paper, the variation of the shape of the feature with the quantity of impurities and the characteristic size and shape of the grains is clearly evidenced.

Due to the various processes used in the samples preparation, differences – sometimes important – can appear at a given wavelength between the measurements of the different authors which are not expected to be fully mutually consistent. Since our purpose is to build a dielectric function as accurate and consistent as possible in the mid-infrared (the IRAS spectrograph covers from 7 to 23μm), it is always chosen to give a greater weight to the more complete measurements in that domain. Practically, we used as basic data the n's and k's given by Phillip and Taft (1960) in the visible and ultraviolet and the absorption spectrum of the GWS sample of SiC 600 measured by Borghesi et al. (1985 and 1986) in the mid- and far-infrared. Among the different samples used in the laboratory experiments, the latter was chosen because it is made of the smallest grains of the purest material. As a consequence, the computations reported here are not relative to a particular sample of silicon carbide but are only to be taken as physically consistent approximations of the optical properties of this important constituent of circumstellar dust.

3. Computation of the dielectric function

In the case of an absorption spectrum of a powder, the quantity measured at a wavelength λ is the extinction efficiency Q_{ext} of the particles, related to the optical index $m = n - ik$ of their constituent material by Mie's theory (Van de Hulst, 1957). When the grain radius a is small in comparison with λ (which is the case in the present study), the absorption and extinction profiles become independent of a and are given by:

$$\frac{Q_{\text{abs}}}{a} \sim \frac{Q_{\text{ext}}}{a} = \frac{2\pi}{\lambda} \cdot \frac{24nk}{(n^2 - k^2 + 2)^2 + 4n^2k^2} \quad (1)$$

if the measurement is performed in vacuum, or:

$$\frac{Q_{\text{abs}}}{a} \sim \frac{Q_{\text{ext}}}{a} = \frac{2\pi}{\lambda} \cdot n_0^3 \frac{24nk}{(n^2 - k^2 + 2n_0^2)^2 + 4n^2k^2} \quad (2)$$

if the grains are embedded in a matrix of index n_0 (Koike et al., 1980).

A supplementary constraint is given by the requirement for the real and imaginary parts of the dielectric function to verify the Kramers-Kronig relation (Landau and Lifshitz, 1960):

$$\varepsilon_1(\omega) = 1 + \frac{2}{\pi} P \int_0^\infty \frac{x \varepsilon_2(x)}{x^2 - \omega^2} \cdot \mathrm{d}x \quad (3)$$

where $\omega = \frac{2\pi c}{\lambda}$, $\varepsilon_1 = n^2 - k^2$, $\varepsilon_2 = 2nk$ and P indicates that the principal value is to be taken.

At very long wavelength, due to the DC conductivity (σ) of silicon carbide, the asymptotic value of the imaginary part of the dielectric function is given by (Landau and Lifshitz, 1960):

$$\varepsilon_2(\lambda \mapsto \infty) = \frac{2\sigma\lambda}{c} = \frac{4\pi\sigma}{\omega} \quad (4)$$

which, substituted in (2), leads to the following behaviours of the Q/a profile:

$$\lambda_c < \lambda < \lambda_c \frac{\varepsilon_1^\infty + 2n_0^2}{\varepsilon_2(\lambda_c)} \mapsto Q/a = \text{constant}$$

$$\lambda > \lambda_c \frac{\varepsilon_1^\infty + 2n_0^2}{\varepsilon_2(\lambda_c)} \mapsto Q/a \propto \lambda^{-2}$$

where ε_1^∞ is the asymptotic value of the real part of the dielectric function and λ_c the critical wavelength beyond which $\varepsilon_2(\lambda)$ is dominated by the contribution of σ. If (as is the case here) no such behaviour is visible on the data, it is only possible to give an upper limit for σ, which is:

$$\sigma_{\text{MAX}} = \frac{c}{2} \left[\frac{\varepsilon_2(\lambda)}{\lambda} \right]_{\lambda = \lambda_{\text{MAX}}} \quad (5)$$

where λ_{MAX} is the maximum wavelength of measurement. However, this uncertainty in the determination of ε_2 for the long wavelengths does not induce any error in the computation of ε_1 since:

$$P \int_0^\infty \frac{dx}{x^2 - \omega^2} = 0$$

Practically, we used the following procedure, analogous to that described in Draine and Lee (1984):

i) ε_2 is chosen over the whole electromagnetic spectrum as close as possible to the existing laboratory measurements;

ii) ε_1 and Q/a are then computed by (3) and (2) and compared with data when available;

iii) where the agreement between the computed and measured values is not satisfactory, the initial values of ε_2 are modified and the two previous steps repeated.

The index n_0 of the KBr matrix was taken in Wolfe and Zissis (1978). Results are displayed in Figs 1 and 2 for the real and imaginary parts of the refractive index of α-SiC. The good agreement between the computed Q/a's and the measurements of Borghesi et al. (1985 and 1986) can be seen in Fig. 3 where the latter are plotted with our computations, both in vacuum (Eq. 1) and in KBr (Eq. 2). Values of the complex refractive index $n + ik$ and of the asymptotic absorption efficiency Q/a in vacuum are also tabulated in Table 1. Taking into account the experimental uncertainties ($\sim 1\%$) given by Borghesi et al. (1985), the accuracy of our results can be estimated to be better than 5%.

4. Discussion

As can be seen in Fig. 1, the general agreement is satisfactory between our computed n's and the measurements of Phillip and Taft and Pikhtin et al. Our results display nevertheless a slightly different behaviour (with a relative difference up to 20%) between 0.2 and 7 μm. This discrepancy is due to the sharp increase of k around 2 μm (Fig. 2), necessary to fit the high values of Q/a measured in that region. The measurements of Pikhtin et al. display a similar – but considerably shallower – elbow on n, confirming the existence of this feature on k for their sample, but

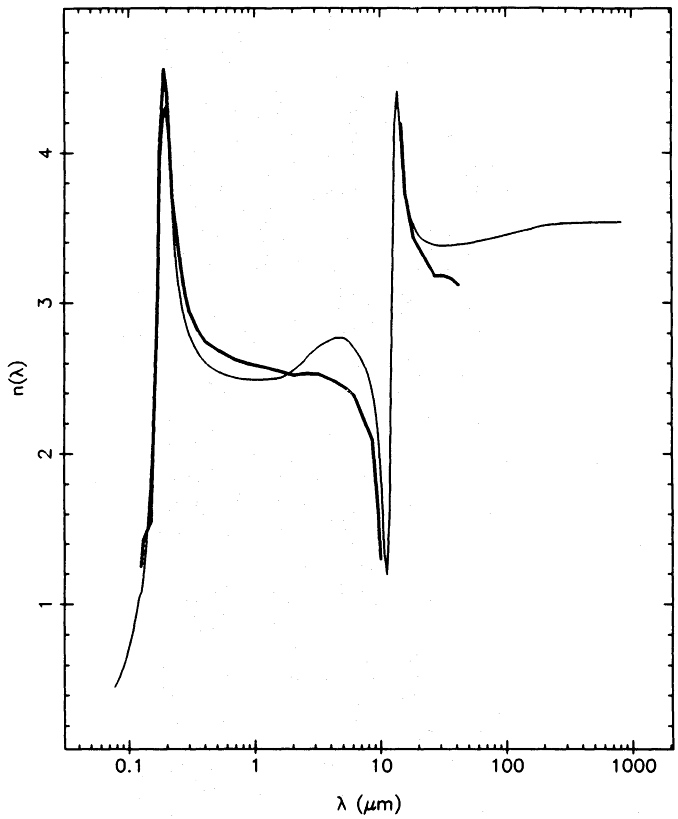

Fig. 1. Real part of the complex refractive index of α-SiC. The thin line is the computations; the thick lines the measurements of Phillip and Taft (1960) below 0.4 μm and that of Pikhtin et al. (1977) for longer wavelengths

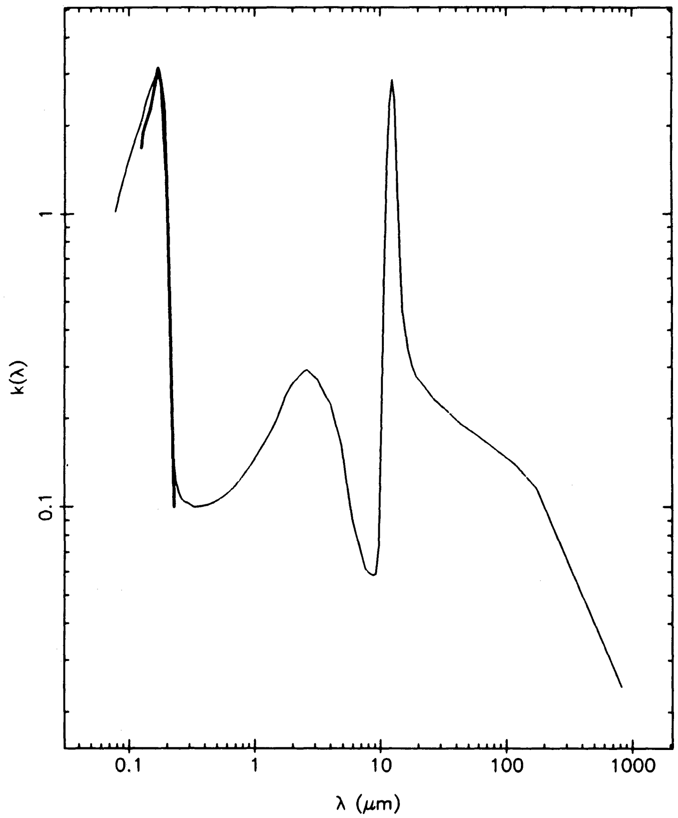

Fig. 2. Imaginary part of the complex refractive index of α-SiC. The thick line is the measurements of Phillip and Taft (1960)

with a quite lower intensity. A possible explanation of this difference is that the measurements of n were performed on pure hexagonal SiC crystals while the absorption spectra were measured on grains made of both hexagonal and rhomboedric SiC. Between 7 and 20 μm, domain of the greatest importance for astrophysical applications, the behaviour of n is dominated by the strong absorption at 11.3 μm and the above discussion on the intensity of the 2 μm bump has only little influence on the determination of the optical properties in this region where the agreement is good. For longer wavelengths, the computed n's are about 10% higher than the measurements. One must be cautious on this last point since the measurements of Borghesi et al. (1985 and 1986) are not perfectly consistent between 20 and 50 μm and since lower Q/a's at long wavelengths would lead to lower n's. These authors do not explain this discrepancy between their two measurements but it must be noticed that a slight difference in the purity or morphology of the material used in these two experiments would be sufficient to produce such a variation of the absorption in the wings of the 11.3 μm feature. As a consequence, we think that our results are the best compromise between the various available measurements and seem to be quite consistent with all of them, except that of Spitzer et al. (1959): as previously noticed by Friedemann et al. (1981), the indices given in this last paper are found to be completely inadequate to reproduce the measured absorption spectra. While the dielectric function of α-SiC seems to be well determined in the vicinity of the 11.3 μm band, further measurements between 0.2 and 7 μm and between 20 and 50 μm would be of great interest and could lead to significative improvements in the optical properties determination in these two regions.

Following the computations of Dorschner et al. (1978), Borghesi et al. (1983, 1985) applied two corrections to their measurements in order to eliminate the influence of the KBr matrix. They are:

i) a wavelength shift $D_\lambda = -0.39$ μm within the range $9 < \lambda < 16$ μm;

ii) a correction factor $h = 0.7 = \frac{Q_{\text{(in vacuum)}}}{Q_{\text{(in KBr)}}}$ over the entire band profile.

A comparison between the two Q/a profiles computed with our final indices in vacuum (Eq. 1) and in KBr (Eq. 2) leads to different values for these two parameters which are $D_\lambda = -0.30$ μm and $h = 0.46$. However, this discrepancy does not seem critical since the values adopted by the previous authors are the average of several determinations the dispersion of which is larger than the difference between our values of D_λ and h and those mentioned above. With these corrections, the peak of maximum absorption is located at 11.33 ± 0.05 μm, i.e. the precise wavelength of maximum intensity of the feature exhibited by the IRAS spectra of class 4n (Baron et al., 1987). This last point strongly supports the assignment of this feature to α-SiC since the available absorption spectra of β-SiC display either a peak

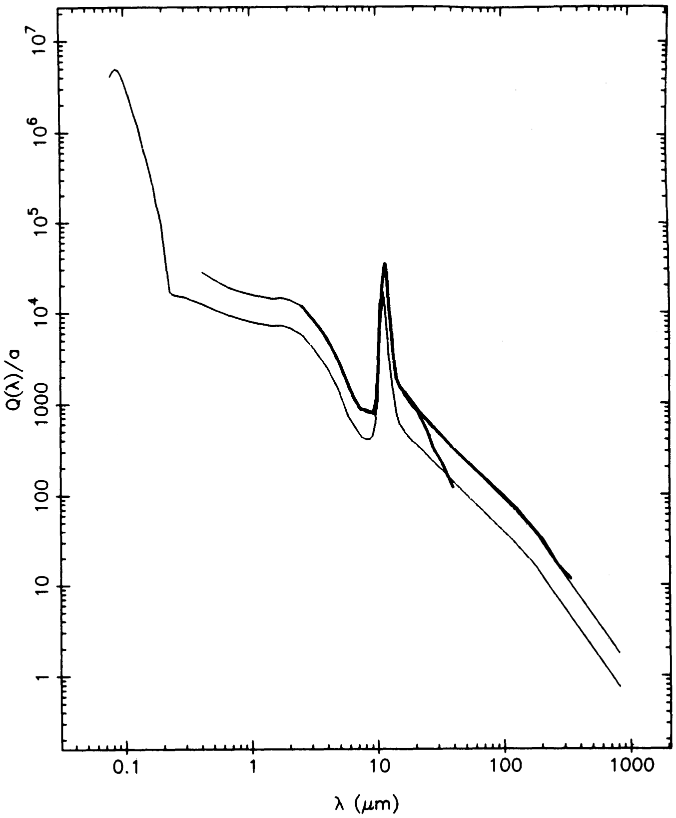

Fig. 3. Computed and measured extinction efficiencies of small grains of α-SiC. The thin lines are computed for particles in vacuum (Eq. 1, lower curve) and in KBr (Eq. 2, upper curve). The thick lines are the experimental data of Borghesi et al. (1985) between 2.5 and 40 μm and of Borghesi et al. (1986) between 20 and 300 μm

Table 1. Complex refractive index and asymptotic extinction efficiency of α-SiC. The Q/a's are computed in vacuum (Eq. 1) for very small particles ($2\pi a/\lambda \ll 1$ at every wavelength)

Wavelength (μm)	n	k	Q/a in vacuum ($cm^{-1}-1$)
0.100	0.706	1.533	$0.346\ 10^7$
0.199	4.250	1.061	$0.765\ 10^5$
0.395	2.617	0.101	$0.129\ 10^5$
0.784	2.496	0.125	$0.885\ 10^4$
0.985	2.489	0.144	$0.813\ 10^4$
1.238	2.492	0.169	$0.760\ 10^4$
1.557	2.500	0.206	$0.729\ 10^4$
1.957	2.544	0.259	$0.701\ 10^4$
2.460	2.614	0.289	$0.587\ 10^4$
3.092	2.698	0.274	$0.415\ 10^4$
3.360	2.722	0.258	$0.353\ 10^4$
3.651	2.739	0.241	$0.300\ 10^4$
3.968	2.755	0.225	$0.255\ 10^4$
4.312	2.767	0.199	$0.206\ 10^4$
4.686	2.769	0.175	$0.166\ 10^4$
5.093	2.767	0.145	$0.127\ 10^4$
5.534	2.747	0.113	$0.928\ 10^3$
6.015	2.713	0.090	$0.697\ 10^3$
6.536	2.672	0.078	$0.577\ 10^3$
7.103	2.621	0.068	$0.483\ 10^3$
7.719	2.551	0.061	$0.419\ 10^3$
8.389	2.455	0.059	$0.403\ 10^3$
9.117	2.289	0.059	$0.424\ 10^3$
9.907	1.930	0.108	$0.965\ 10^3$
10.137	1.792	0.170	$0.166\ 10^4$
10.371	1.600	0.279	$0.306\ 10^4$
10.611	1.409	0.462	$0.576\ 10^4$
10.857	1.296	0.732	$0.935\ 10^4$
11.108	1.230	1.126	$0.137\ 10^5$
11.365	1.281	1.577	$0.145\ 10^5$
11.628	1.488	1.981	$0.104\ 10^5$
11.897	1.803	2.385	$0.730\ 10^4$
12.172	2.383	2.634	$0.484\ 10^4$
12.454	3.076	2.813	$0.331\ 10^4$
12.742	3.607	2.619	$0.253\ 10^4$
13.037	4.194	2.401	$0.194\ 10^4$
13.339	4.294	1.734	$0.151\ 10^4$
13.647	4.396	1.253	$0.118\ 10^4$
13.963	4.298	0.948	$0.969\ 10^3$
14.286	4.188	0.719	$0.796\ 10^3$
14.617	4.067	0.587	$0.695\ 10^3$
14.955	3.946	0.488	$0.615\ 10^3$
15.301	3.853	0.438	$0.576\ 10^3$
15.655	3.774	0.409	$0.555\ 10^3$
18.585	3.503	0.289	$0.400\ 10^3$
22.063	3.415	0.260	$0.323\ 10^3$
26.193	3.387	0.236	$0.252\ 10^3$
31.095	3.377	0.220	$0.199\ 10^3$
36.915	3.380	0.205	$0.157\ 10^3$
43.824	3.386	0.192	$0.123\ 10^3$
52.026	3.396	0.182	$0.974\ 10^2$
61.763	3.408	0.172	$0.772\ 10^2$
73.323	3.422	0.164	$0.611\ 10^2$
87.046	3.437	0.155	$0.481\ 10^2$
103.338	3.452	0.146	$0.379\ 10^2$
122.679	3.468	0.137	$0.297\ 10^2$
145.639	3.483	0.126	$0.228\ 10^2$
172.897	3.499	0.116	$0.174\ 10^2$
205.257	3.513	0.098	$0.123\ 10^2$
243.673	3.520	0.083	$0.869\ 10^1$

located at shorter wavelength (Borghesi et al., 1985) or even a double-peaked profile (Stephens, 1980).

The Q/a profiles of Borghesi et al. (1986) do not exhibit any shoulder characterizing σ for wavelengths as long as $\lambda_{MAX} = 300\ \mu m$. This corresponds (Eq. 5) to a resistivity $1/\sigma_{MAX} > 1.2\Omega$ cm, well consistent with the values given by Pikhtin et al. (1977) who indicate that the resistivity of very pure SiC crystals – at room temperature – lies between 3 and 10Ω cm.

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