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ON SILICON CARBIDE GRAINS AS THE CARRIER OF THE 21 μm EMISSION FEATURE IN POST–ASYMPTOTIC GIANT BRANCH STARS

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ABSTRACT

The mysterious 21 μm emission feature seen in 12 proto–planetary nebulae remains unidentified since its first detection in 1989. Over a dozen candidate materials have been proposed within the past decade, but none of them have received general acceptance. Very recently, silicon carbide (SiC) grains with impurities were suggested to be the carrier of this enigmatic feature, based on recent laboratory data that doped SiC grains exhibit a resonance at ~21 μm. This proposal gains strength from the fact that SiC is a common dust species in carbon-rich circumstellar envelopes. However, SiC dust has a strong vibrational band at ~11.3 μm. We show in this Letter that in order to be consistent with the observed flux ratios of the 11.3 μm feature to the 21 μm feature, the band strength of the 21 μm resonance has to be very strong, too strong to be consistent with current laboratory measurements. But this does not yet readily rule out the SiC hypothesis since recent experimental results have demonstrated that the 21 μm resonance of doped SiC becomes stronger as the C impurity increases. Further laboratory measurements of SiC dust with high fractions of C impurity are urgently needed to test the hypothesis of SiC as the carrier of the 21 μm feature.

Subject headings: circumstellar matter — dust, extinction — infrared: stars — stars: AGB and post-AGB — stars: individual (HD 56126)

1. INTRODUCTION

The so-called 21 μm feature has been identified in 12 proto–planetary nebulae (PPNe; Kwok et al. 1999)—and arguably also in two planetary nebulae associated with Wolf-Rayet central stars (Hony et al. 2001) and in two highly evolved carbon stars (Volk et al. 2000)—since its first detection in 1989 (Kwok et al. 1989). This feature has a similar spectral shape and peaks at the same wavelength (~20.1 μm) in all sources. The 21 μm feature sources have quite uniform properties; they are mostly metal-poor, carbon-rich F and G supergiants with infrared excesses and overabundant *s*-process elements (see Kwok et al. 1999).

The origin of this feature, however, is still a mystery. A large number of candidate carriers have been proposed in the past decade, including hydrogenated fullerenes, polycyclic aromatic hydrocarbons (PAHs), hydrogenated amorphous carbons, diamonds, synthetic carbonaceous macromolecules, amides (thiourea or urea $OC[NH_2]_2$), iron oxides (such as Fe_2O_3 or Fe_3O_4), SiS_2, and oxygen-bearing side groups in coal (see Kwok et al. 1999, Andersen et al. 2005, and references therein) and, more recently, titanium carbide (TiC) nanoclusters (von Helden et al. 2000), stochastically heated silicon core-SiO_2 mantle nanograins (Smith & Witt 2002; Li & Draine 2002), doped SiC (Speck & Hofmeister 2004), SiC core-SiO_2 mantle grains, and iron monoxide FeO (Posch et al. 2004).

Among these candidates, TiC nanograins have recently received much attention, because (1) laboratory spectra of TiC nanocrystals exhibits a distinct feature at ~20.1 μm, closely resembling the astronomical 21 μm emission feature both in peak position and width and in spectral details (von Helden et al. 2000), although bulk TiC does not show any noticeable feature near 20.1 μm (Henning & Mutschke 2001), and (2) presolar TiC grains are identified in primitive meteorites as nanometer-sized inclusions embedded in micrometer-sized presolar graphite grains (Bernatowicz et al. 1996). However, the TiC model has been challenged by Hony et al. (2003), Chigai et al. (2003), and Li (2003).[3]

More recently, SiC seems to be an attractive candidate for the 21 μm feature carrier: Speck & Hofmeister (2004) reported the experimental finding that SiC, under certain circumstances, not only shows the well-known resonance feature at ~11.3 μm but also a secondary band that is centered at 20–21 μm. This secondary band was reported to appear only in the β SiC polytype, and nitrogen or carbon impurities were suspected to favor its occurrence. This hypothesis gains strength from the fact that (1) both silicon and carbon are abundant elements, and (2) SiC is a common dust species in C-rich circumstellar envelopes. Due to the astrophysical significance of SiC dust, it deserves a more thorough investigation. In this Letter, we aim at investigating whether or not doped SiC can be a suitable carrier for the mysterious 21 μm feature by comparing the model-predicted flux ratio of the 21 μm feature to the 11.3 μm feature with the observed flux ratios.

2. CIRCUMSTELLAR SiC GRAINS AND THEIR OPTICAL PROPERTIES

As early as 1933, Wildt had already suggested that SiC grains might form in the cool atmospheres of N-type stars (Wildt 1933). This suggestion was confirmed 36 years later when Gilman (1969) and Friedemann (1969) performed thermodynamical equilibrium

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[3] Hony et al. (2003) found that nano-TiC must absorb much more strongly in the optical and ultraviolet (UV) wavelengths than its bulk counterpart in order for the required amount of TiC dust not to exceed the (observed) maximum available Ti abundance. Chigai et al. (2003) found that in order to be consistent with the observed flux ratio of the 21 μm band to the 11.3 μm band, the Ti/Si abundance ratio must be at least 5 times larger than the solar abundance ratio. One may argue that the arguments of Hony et al. (2003) and Chigai et al. (2003) do not readily rule out the TiC hypothesis because (1) as a consequence of the so-called *electron mean free path limitation* effect, the imaginary parts of the dielectric functions of small metallic grains (and therefore their optical/UV absorptivities) are expected to be larger than those of their bulk counterparts (see Li 2004) and (2) there is no reason to compare the solar Ti/Si abundance ratio with that of the 21 μm feature sources. However, applying the Kramers-Kronig physical principle to the TiC hypothesis, Li (2003) readily ruled out the TiC model because it was found that this model requires at least 50 times more TiC mass than available, no matter how strong the UV/optical absorptivity of nano-TiC is.

calculations and found that SiC grains could condense in carbon stars. The presence of circumstellar SiC grains was first revealed by the detection of an emission feature at 11.5 μm in some carbon stars that was first attributed to SiC dust by Gilra (1972). The detection of this feature and its identification as SiC were confirmed by subsequent observations and interpretations (Treffers & Cohen 1974; Merrill & Stein 1976; Goebel et al. 1980; Little-Marenin 1986; Goebel et al. 1995; Speck et al. 1997). The formation of SiC dust in carbon stars has also been indicated by the identification of presolar SiC grains in primitive meteorites based on their isotopic anomalies (Bernatowicz et al. 1987).

SiC has ~70 polytypes, which can be divided into two general crystallographic types: cubic (β SiC) and hexagonal (α SiC). While presolar SiC grains were predominantly found to have a cubic lattice structure (i.e., β SiC; Daulton et al. 2003), the 11.3 μm emission feature observed for carbon-rich asymptotic giant branch (AGB) stars is best fitted by laboratory spectra of α SiC (Baron et al. 1987). Speck et al. (1999) suggested that this discrepancy (between meteoritic and astronomical identifications of the SiC type) is caused by the "inappropriate 'KBr corrections' having been made to laboratory spectra of SiC particles dispersed in KBr matrices"; they argued that the carrier of the 11.3 μm feature seen in carbon stars is actually β SiC (instead of α SiC). More recently, Clément et al. (2003) found that the laboratory spectra of matrix-isolated β SiC nanoparticles perfectly match the astronomical 11.3 μm feature. But Mutschke et al. (1999) argued that the 11.3 μm emission feature itself is not a powerful discriminator of the SiC crystal type (also see Papoular et al. 1998).

Therefore, in this work we will consider both types of SiC grains. We approximate the grains as spherical[4] and use Mie theory to calculate their absorption and scattering properties. The complex index of refraction $m(\lambda) = m'(\lambda) + im''(\lambda)$ of α SiC is taken from Laor & Draine (1993).[5] For β SiC, Adachi (1999) compiled the refractive indices recently determined by various groups in the wavelength range of $0.13\ \mu\mathrm{m} \le \lambda \le 124\ \mu\mathrm{m}$, except there were no m'' data for $0.5\ \mu\mathrm{m} \le \lambda \le 0.65\ \mu\mathrm{m}$. Largely based on the data compiled by Adachi (1999), we take the following "synthetic" approach: for $\lambda \le 0.13\ \mu\mathrm{m}$, we take the imaginary parts of the refractive indices m'' from Laor & Draine (1993);[6] for $0.13\ \mu\mathrm{m} \le \lambda \le 0.5\ \mu\mathrm{m}$ and $0.65\ \mu\mathrm{m} \le \lambda \le 124\ \mu\mathrm{m}$, we take m'' from Adachi (1999); for $0.5\ \mu\mathrm{m} \le \lambda \le 0.65\ \mu\mathrm{m}$, extrapolation is made from that of Adachi (1999) at $\lambda \le 0.5\ \mu\mathrm{m}$; for $\lambda > 124\ \mu\mathrm{m}$, we assume $m''(\lambda) = m''(124\ \mu\mathrm{m})(124\ \mu\mathrm{m}/\lambda)$. After smoothly joining the adopted m'', we calculate the real part $m'(\lambda)$ from m'' through the Kramers-Kronig relation (Bohren & Huffman 1983).

If SiC grains are indeed the carrier of the observed 21 μm feature, they must have a resonance at this wavelength, and we should include its contribution to their dielectric functions. We approximate this contribution in terms of a single Lorentz oscillator:

$$\delta\epsilon = \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma\omega}, \qquad (1)$$

where $\omega \equiv 2\pi c/\lambda$ is the angular frequency (c is the speed of light), ω_0 is the angular frequency of the transverse optical mode, $\gamma = 2\pi c\Delta\lambda_0/\lambda_0^2$ is the damping constant ($\lambda_0 \approx 20.1\ \mu$m and $\Delta\lambda_0 \approx 2\ \mu$m are, respectively, the peak wavelength and the FWHM of the 21 μm feature), and ω_p is the plasma frequency. For spherical grains in the Rayleigh regime, the parameters ω_p and ω_0 can be determined from the feature strength ($Q_{\rm abs}/a$) (Bohren & Huffman 1983):[7]

$$\omega_p = \frac{1}{3}\left[\left(\frac{Q_{\rm abs}}{a}\right)\frac{3\gamma c}{4}\right]^{1/2}[\epsilon(\infty) + 2], \qquad (2)$$

$$\omega_0 = \left[\left(\frac{2\pi c}{\lambda_0}\right)^2 - \frac{\omega_p^2}{\epsilon(\infty) + 2}\right]^{1/2}, \qquad (3)$$

where $\epsilon(\infty)$ is the dielectric function at $\omega \to \infty$: $\epsilon(\infty) \approx 6.6(6.7)$ for α SiC (β SiC).

Unfortunately, Speck & Hofmeister (2004) did not measure the absolute strength of the 21 μm feature ($Q_{\rm abs}/a$) for their SiC samples. We therefore treat ($Q_{\rm abs}/a$) as a free parameter: for a given ($Q_{\rm abs}/a$) value, we calculate the contribution of the 21 μm feature to the dielectric functions of SiC from equations (1), (2), and (3) and add this component to the dielectric functions of Laor & Draine (1993) for α SiC and those described early in this section for β SiC.[8] For illustrative purpose, in Figure 1 we plot the refractive indices of α SiC and β SiC with $(Q_{\rm abs}/a) = 0, 100, 10^3, 10^4\ \mathrm{cm}^{-1}$.

3. RESULTS

The 21 μm sources all have a weak emission feature at 11.3 μm, which is commonly attributed to PAHs (see Kwok et al. 1999). Assuming that all the power from the 11.3 μm feature is emitted by SiC grains, one can place constraints on the size and the 21 μm feature strength ($Q_{\rm abs}/a$) of SiC dust by comparing the power emitted from the 11.3 μm feature with that emitted from the 21 μm feature. For this purpose, we take HD 56126 (for which the dust and gas spatial distributions are well constrained; see Hony et al. 2003, Meixner et al. 2004, and Hrivnak & Bieging 2005) as a testing example.

HD 56126 (IRAS 07134+1005), a bright post-AGB star with a spectral type of F0–5 I, is one of the four 21 μm sources originally discovered by Kwok et al. (1989) and remains the best-studied 21 μm source. The total power emitted from the 11.3 μm feature and from the 21 μm feature are, respectively, $E(11.3\ \mu\mathrm{m}) \approx 1.8 \times 10^{-11}$ and $E(21\ \mu\mathrm{m}) \approx 1.5 \times 10^{-9}$ ergs $\mathrm{s}^{-1}\ \mathrm{cm}^{-2}$ (Hony et al. 2003). Now the question is, with what grain size and what ($Q_{\rm abs}/a$) for the 21 μm feature can one achieve $E(11.3\ \mu\mathrm{m})/E(21\ \mu\mathrm{m}) < 0.012$?

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[4] If the grain shape is approximated as a continuous distribution of ellipsoids (Bohren & Huffman 1983), both the 11.3 μm feature and the 21 μm feature will be significantly broadened. But since at these wavelength ranges, micron or submicron-sized SiC grains are in the Rayleigh regime, the broadening effect is unlikely to differ much from one feature to another. Therefore, it is sufficient to just consider spherical grains.

[5] Laor & Draine (1993) based their dielectric functions ($\epsilon = m^2$) in the 11.3 μm wavelength range on those of Spitzer et al. (1959), but broadened by a factor of ~16.7. In addition, they introduced a "continuum" by adding a highly damped oscillator.

[6] At such short wavelengths, the dielectric functions for α SiC and β SiC should not differ much since they just depend on the atomic absorption cross sections of the constituent atoms and are not sensitive to the exact crystal structure.

[7] $Q_{\rm abs}$ is the absorption efficiency at $\lambda_0 = 20.1\ \mu$m, and a is the grain radius. For grains in the Rayleigh approximation (i.e., $2\pi a/\lambda \ll 1$), $(Q_{\rm abs}/a) \equiv (4\rho/3)\,\kappa_{\rm abs}$ is independent of a, where $\rho \approx 3.22\ \mathrm{g\ cm^{-3}}$ is the mass density of SiC and $\kappa_{\rm abs}$ is the mass absorption coefficient of SiC at 20.1 μm. In this work, $Q_{\rm abs}/a$ is treated as a free parameter.

[8] The Lorentz oscillator nature of the 21 μm resonance (see eq. [1]) guarantees that the new dielectric functions also satisfy the Kramers-Kronig relation (see Bohren & Huffman 1983).

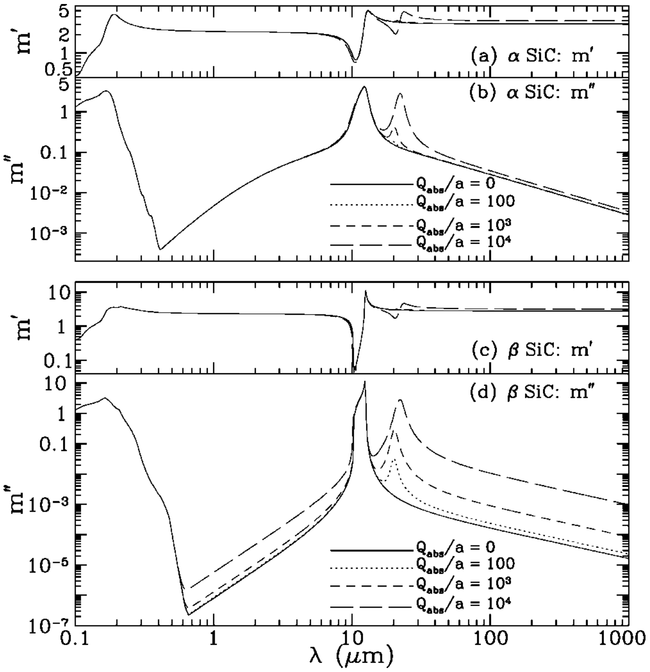

FIG. 1.—Refractive indices $m(\lambda) = m'(\lambda) + im''(\lambda)$ of (*a*, *b*) α SiC and (*c*, *d*) β SiC with various strengths for the 21 μm feature: $Q_{\rm abs}/a = 0$ (*solid lines*), $Q_{\rm abs}/a = 100$ cm^{-1} (*dotted lines*), $Q_{\rm abs}/a = 10^3$ cm^{-1} (*short-dashed lines*), and $Q_{\rm abs}/a = 10^4$ cm^{-1} (*long-dashed lines*).

Apparently, the observational requirement of $E(11.3\ \mu{\rm m})/E(21\ \mu{\rm m}) < 0.012$ is best met if the grains are cold (say, with an equilibrium temperature ~150 K) and have a large 21 μm feature strength ($Q_{\rm abs}/a$). Since the farther the grains are away from the central illuminating star, the colder the grains are, we just need to consider how cold SiC dust can be at the outer edge of the dusty envelope around HD 56126. On the other hand, ($Q_{\rm abs}/a$) for the 21 μm feature cannot be arbitrarily large—it should not be inconsistent with laboratory measurements (e.g., see Fig. 1 in Speck & Hofmeister 2004).

The equilibrium temperature of a SiC grain of spherical radius a at the outer edge of the dust envelope around HD 56126 can be determined by balancing absorption and emission:

$$\left(\frac{R_\star}{2r_{\rm max}}\right)^2 \int_0^\infty C_{\rm abs}(a,\lambda)F_\lambda^\star \exp(-A_\lambda/1.086)d\lambda = \int_0^\infty C_{\rm abs}(a,\lambda)4\pi B_\lambda(T[a])d\lambda, \qquad (4)$$

where $R_\star \approx 49.2\ R_\odot$ is the stellar radius ($R_\odot$ is the solar radius); $r_{\rm max} \approx 9.3 \times 10^{16}$ cm is the distance from the central star to the outer edge of the dust envelope (Hony et al. 2003); $C_{\rm abs}(a, \lambda)$ is the absorption cross section of SiC dust of size a at wavelength λ; $T(a)$ is the equilibrium temperature of dust of size a at $r_{\rm max}$; $F_\lambda^\star$ is the flux per unit wavelength (ergs s^{-1} cm^{-2} μm^{-1}) at the top of the illuminating star's atmosphere, which is approximated by the Kurucz (1979) model atmospheric spectrum with $T_{\rm eff} = 7250$ K and $\log g = 1.0$; and A_λ is the dust extinction that is represented by the Milky Way standard extinction law with $A_V = 1.0$ mag (Hony et al. 2003).[9]

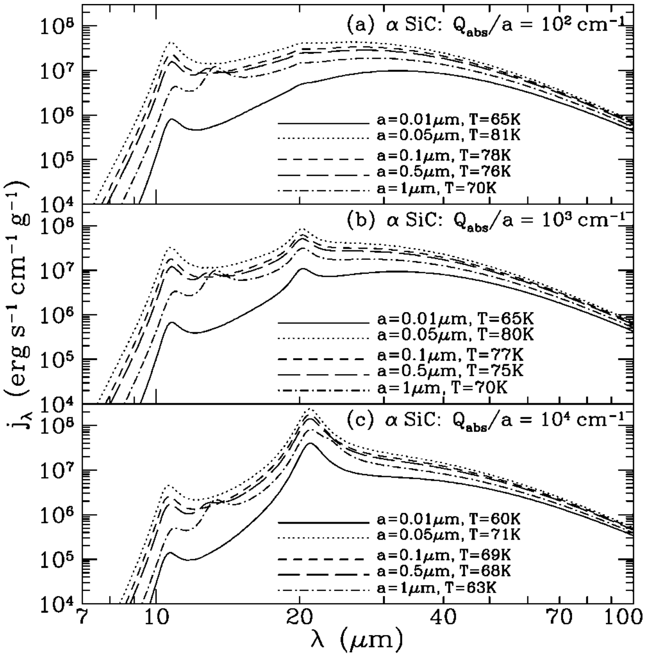

FIG. 2.—Emission spectra for α SiC grains of sizes $a = 0.01$ μm (*solid lines*), $a = 0.05$ μm (*dotted lines*), $a = 0.1$ μm (*short-dashed lines*), $a = 0.5$ μm (*long-dashed lines*), and $a = 1.0$ μm (*dot-dashed lines*), with (*a*) ($Q_{\rm abs}/a$) = 100 cm^{-1}, (*b*) ($Q_{\rm abs}/a$) = 10^3 cm^{-1}, and (*c*) ($Q_{\rm abs}/a$) = 10^4 cm^{-1} for the 21 μm feature. The grains are at the outer edge of the dust envelope around HD 56126.

For a SiC grain of a given size a and a given 21 μm feature strength ($Q_{\rm abs}/a$), we calculate its equilibrium temperature from equation (4) and its emission spectrum and then calculate $E(11.3\ \mu{\rm m})/E(21\ \mu{\rm m})$—the ratio of the amount of energy emitted from the 11.3 μm feature to that from the 21 μm feature.

In Figure 2 we plot the emission spectra of α SiC grains of sizes $a = 0.01$, 0.05, 0.1, 0.5, and 1.0 μm with ($Q_{\rm abs}/a$) = 100, 1000, 10^4 cm^{-1} for the 21 μm feature. It is seen that even if one assumes ($Q_{\rm abs}/a$) = 10^4 cm^{-1}, the calculated $E(11.3\ \mu{\rm m})/E(21\ \mu{\rm m})$ ratio (≈ 0.61 for $a = 0.01$ μm and ≈ 0.41 for $a = 1.0$ μm) is still much larger than the observed ratio of $E(11.3\ \mu{\rm m})/E(21\ \mu{\rm m}) < 0.012$.[10] In order to be consistent with the observation, one requires ($Q_{\rm abs}/a$) $\gg 10^4$ cm^{-1}. This seems unlikely since the required strength for the 21 μm feature is stronger than that for the 11.3 μm feature of α SiC, which is only ($Q_{\rm abs}/a$) $\approx 1.5 \times 10^4$ cm^{-1}.[11] Similar results are obtained for β SiC (see Fig. 3).[12]

4. DISCUSSION

In § 3 we only consider grains large enough to attain an equilibrium temperature. For small grains with $a < 100$ Å, they will undergo single-photon heating: upon absorption of an energetic photon, they will be heated to a temperature that is higher than their equilibrium temperature, and most of the pho-

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[9] This extinction correction is not exact, but this does not affect our results since we only need to approximately estimate the amount of dust attenuation (see Fig. 1 of Hony et al. 2003).

[10] Note that $E(11.3\ \mu{\rm m})/E(21\ \mu{\rm m}) < 0.012$ is already a very generous upper limit since we have attributed the entire 11.3 μm emission to SiC, while actually this emission must at least partly originate from PAHs.

[11] As a matter of fact, in the experimental spectra of the β SiC samples of Speck & Hofmeister (2004), the 21 μm resonance is far weaker than the 11.3 μm resonance for both bulk materials and nanomaterials.

[12] In addition to the prominent 11.3 μm band, β SiC appears to have another sharp band at ~12.6 μm. This is caused by the asymmetrical nature of its index of refraction (Adachi 1999; see Fig. 1*d*). If we approximate the 11.3 μm resonance by a Lorentz oscillator, the secondary peak at ~12.6 μm would disappear. But this does not affect our conclusion since we are considering the total power emitted in these features, rather than their peak heights.

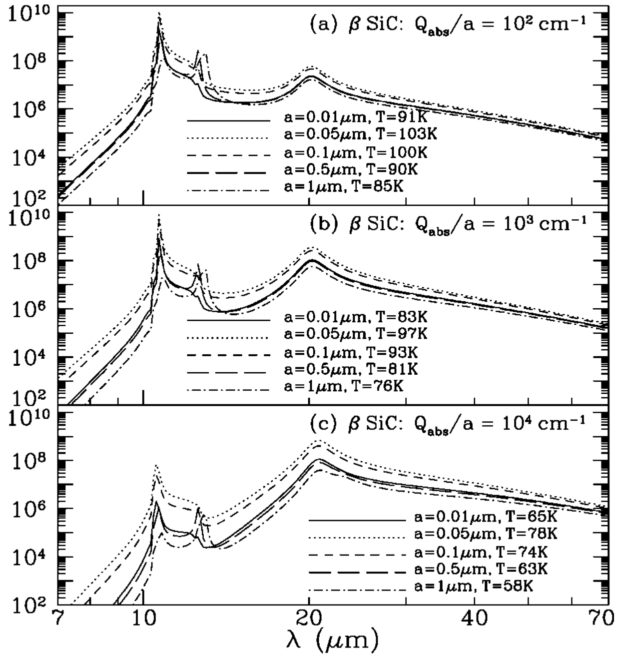

FIG. 3.—Same as Fig. 2, but for β SiC

ton energy will be radiated away at this high temperature. Therefore, the emission spectrum of a stochastically heated grain peaks at shorter wavelengths than that calculated from its equilibrium temperature (see Draine & Li 2001). We thus would expect larger ratios of $E(11.3\ \mu m)/E(21\ \mu m)$ for small grains undergoing single-photon heating than those considered above, making things even worse.

However, it is still premature to rule out the SiC hypothesis since the mid-IR spectra of doped SiC samples of Speck & Hofmeister (2004) were obtained at *room temperature* for SiC with a limited fraction of C impurity. While it is unclear how and to what degree temperatures will affect the 11.3 and 21 μm features of SiC, it is known that the mid-IR spectra of silicates are affected by the sample temperature (see Bowey et al. 2001). Moreover, recent experimental results have shown that the strength of the 21 μm feature increases as the impurity content increases (Kimura et al. 2005a, 2005b). The required 21 μm feature strength (see § 3) might be attainable by SiC dust with a rather high level of impurity. Laboratory measurements of the mid-IR spectra of heavily doped SiC samples are urgently needed.

Finally, we note that there are over 700 C-rich AGB stars known to show the 11.3 μm SiC feature from the *Infrared Astronomical Satellite* Low Resolution Spectrometer survey, but none of these stars show the 21 μm feature, suggesting that the amount of impurity incorporated into SiC dust must be dependent on environment (e.g., the Si/C ratio; Speck & Hofmeister 2004), if doped SiC is indeed the carrier of the 21 μm feature.

In summary, we have examined the recent hypothesis of doped SiC as the carrier of the mysterious 21 μm emission feature detected in 12 PPNe. It is found that doped SiC grains have to have a resonance at ~21 μm, too strong to be consistent with current laboratory measurements, in order for the model-predicted flux ratios of the 11.3 μm feature to the 21 μm feature not to be in conflict with the observed values. Admittedly, it is still premature to discard the SiC hypothesis since recent experimental results have shown that the strength of the 21 μm resonance of doped SiC appears to increase with the C impurity content, suggesting that heavily doped SiC may be able to produce a sufficiently strong 21 μm resonance. We call on laboratory measurements of the mid-IR spectra of SiC with high levels of C impurity.

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