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On the crystalline silicate component of comet dust

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Abstract. The effects of preaggregation silicate crystallinity and postaggregation silicate crystallinity on the $11.3 \mu\text{m}$ structure in comet dust emission have been calculated. Of the order of 10 – 20% crystallinity provides the best agreement to date with observation but the fits are not yet fully satisfactory.

1. Introduction

The $10 \mu\text{m}$ feature in the emission by comet dust which was first detected and identified by Ney (Maas, Ney & Woolf 1970) has now been observed in four comets to consist of two main features: one at about $9.7 \mu\text{m}$ and the other at about $11.3 \mu\text{m}$ (see Hanner et al 1994a for a review). The former is generally attributed to the amorphous form of olivine and the latter to the crystalline form of olivine. The latter identification is not as secure as the first and has also been variously attributed to a PAH (Polycyclic Aromatic Hydrocarbon) emission or SiC. If comets were formed out of the cold interstellar dust as proposed by Greenberg and others (Greenberg 1982; Yamamoto 1995) and the interstellar dust contained only amorphous silicates as indicated by its featureless $10 \mu\text{m}$ absorption, what is the source of crystallinity?

If there is some crystalline fraction in the silicates, there are essentially two possibilities for its origin: 1) before the comet formed, some of the interstellar dust was heated to the point at which conversion from amorphous to crystalline silicate occurred; 2) the conversion from amorphous to crystalline occurs after the dust comes off the comet.

Since amorphous silicates crystallize at 875 K in 105 hr (Koike & Tsuchiyama 1992) while at 1000 K the process is essentially instantaneous (Day & Donn 1978), it is understandable that the interstellar dust could have been heated to the required temperature in the protosolar nebula. On the other hand the comet dust could never have achieved such high temperatures at the solar distances at which the $11.3 \mu\text{m}$ structure is already observed. For comet Mueller 1993a (Hanner et al 1994b) the $11.3 \mu\text{m}$ feature appears at a distance of 2 AU. At this distance, the temperature of the fluffy comet dust in a McDonnell size distribution which could provide silicate emission excesses ($m \leq 10^{-8}g$) would be in the range $320 \leq T_d \leq 500\text{K}$ (Greenberg & Hage 1990; Greenberg & Li 1996). How the silicate in the comet dust can be partially metamorphosed turns out

to be a somewhat subtler mechanism than direct *total* heating but on the other hand, it provides a possibility for arriving at a picture more consistent with the cold interstellar dust aggregation of comets whereas the preheating of the dust leads to some inconsistencies with current understanding of the comet nucleus composition with regard to both volatiles and degree of ice crystallinity.

In section 2 we present a summary of the chemical and morphological structure of interstellar dust based on recent observations and interpretations. The third section is a brief discussion of the consequences to the comet nucleus of heating and recondensation of a significant fraction of the interstellar dust in the protosolar nebula. In section 4 we present some introductory remarks on a possible mechanism for converting a fraction of the interstellar silicate to crystallinity by heating to temperatures far below 800 K. Section 5 contains some calculations of the spectral emission by comet dust consisting of fluffy aggregates of partially modified interstellar dust minus their volatile components.

2. Interstellar dust components

The major emphasis in the paper is to follow the refractory component of interstellar dust and in comets.

2.1. Interstellar silicate

The most clearly demonstrated ubiquitous solid material in interstellar space is concluded from its 10 μm Si-O stretch absorption to be a silicate. The optical constants of crystalline olivine — or any other crystalline silicate — provide much sharper features than those observed in the interstellar medium so that it is generally accepted that the interstellar silicates are amorphous. The most clearly defined properties of the interstellar silicates can be deduced better from polarization than from absorption because the latter requires a knowledge of the background as well as possible silicate emission. The polarization observed for the Becklin-Neugebauer object at both 9.7 μm and 18 μm provides the best basis for comparison with dust models. It can be shown that no pure silicates which have been proposed provide a fully suitable match to the observation (Greenberg & Li 1995). However, if an “astronomical silicate” is defined by the measured properties of an amorphous (glassy) silicate core with organic refractory mantle (see section 2.2) a remarkably good fit with the observations is possible (Greenberg & Li 1995). Since the 3.4 μm feature of organics is extremely well correlated with the 9.7 μm absorption (Pendleton et al 1994) it appears that the silicates are indeed coated by an organic refractory mantle so that no (or little) purely silicate particles exist in the interstellar medium. Thus the term “astronomical silicate” is taken to refer to a silicate core-organic refractory mantle particle. The optical constants of the amorphous silicate MgFeSiO_4 are given in Dorschner et al (1995).

2.2. Organic refractory

The presence of a carbonaceous component in interstellar dust that exhibits the 3.4 μm features characteristic of CH stretches in CH_2 and CH_3 groups is now well established (Butchart et al 1986; Sandford et al 1991). Although many

carbon compounds exhibit features similar to the interstellar one, a recent survey (Pendleton et al 1994) shows that no previously produced laboratory analog candidate absorbs exactly like the interstellar dust. A simulation of cyclic interstellar dust evolution which occurs sequentially in molecular and diffuse clouds is accomplished in two stages. The analog of the cyclic ultraviolet processing of interstellar dust described in the following provides a remarkably close match to the observed $3.4 \mu m$ absorption. In the first stage we produce laboratory residues, which result from ultraviolet photoprocessing of analog grain mantles that start out as simple ices (H_2O , CO , CH_4 , CH_3OH , NH_3 , etc.) at 10 K (Greenberg 1978). These "first generation" organics are then exposed to long-term ultraviolet radiation by exposure to the Sun for the full time available on the EURECA satellite ERA platform (Innocenti & Mesland 1995).

The evolution of interstellar dust organics resulting from extensive exposure in diffuse clouds to ultraviolet irradiation of organic mantles produced in interstellar clouds is well simulated by the long-term ultraviolet irradiation of laboratory organic residues in the solar environment. The resulting $3.4 \mu m$ infrared feature of the solar irradiated laboratory organic residues is much closer to that of diffuse cloud interstellar dust than that of a wide variety of other suggested sources of organics (Greenberg et al 1995). In Fig.1 we present a full infrared spectrum of the solar irradiated organics. Note particularly the absorption at $\lambda \geq 5 \mu m$ which plays an important role in modifying the $9.7 \mu m$ feature of pure silicates (Greenberg & Li 1995). For the interstellar case, the absorptivity beyond $5 \mu m$ is probably smaller than that for the ERA sample, but for comet organics it could be as high or higher than the ERA values because the organics made in the molecular cloud contraction leading to comets and solar system have not been exposed to the degree of ultraviolet radiation of the organic refractory mantle as in the diffuse cloud stage. The absorptivities of the organic refractory mantle in the visual and near ultraviolet are responsible for the heating of the comet dust particles as originally noted by Greenberg & Hage (1990). In the current calculation we use $m'' = 0.15$ in the visual as in the earlier work (Greenberg & Hage 1990). We note that this is probably an underestimate for processed organics (Jenniskens 1993).

3. Heating of interstellar dust in the protosolar nebula

Let us assume that the material out of which the comet nuclei formed was initially distributed throughout the protosolar nebula. In other words some of the interstellar dust was modified by heating to various degrees. As an approximation we shall assume that the early Sun was similar in its radiation output to the current one so that the temperature of the dust at various solar distances can be calculated assuming the present Sun radiation. Using the silicate core-organic refractory mantle model of presolar interstellar dust as the basic refractory component the temperature of ~ 875 K at which amorphous silicates are crystallized implies that the organic refractory component is at least partially evaporated and the volatile (ices) components are fully evaporated.

It will turn out that, at least 15% of the silicates in comet dust must be crystallized in order to account for the $11.3 \mu m$ feature. If this crystalline silicate is to be recirculated out to the region where comet formation takes

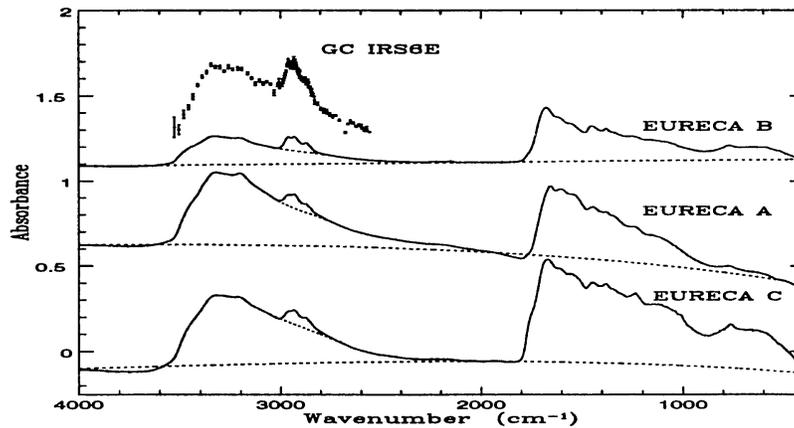


Figure 1. Comparison of the absorbance of three solar irradiated residues on the EURECA satellite ERA platform (Greenberg et al 1995) with the Galactic center GC IRS 6E (Pendleton et al 1994).

place two factors must be taken into account in the composition of the nucleus aggregation: 1) recondensation of volatiles on the silicates; 2) chemical processes in the protosolar nebula. We first note that, if even only 15% of the silicate achieves a temperature of 875 K a much larger fraction of the dust must have been raised to temperatures at which all the ices have evaporated and even some organic mantles as well. In order to estimate what fraction of ices have been evaporated, we first assume a radial distribution of the dust and let the number of dust particles which have achieved $T \geq 875\text{K}$ be 15% of the total. We then conservatively let the fractional number of dust particles with $T \geq 300\text{K}$ be those for which not only the H_2O but all volatiles have been evaporated. The H_2O by itself would sublime at a much lower temperature of $\sim 180\text{K}$ (Hanner 1981), so that the following estimate of the degree of recondensation is a lower limit. If the total ensemble of dust grains then aggregates to form the comet nucleus the fraction of recondensed to initial ice is greater than the ratio of the numbers with $T \geq 300\text{K}$ to those with $T \leq 300\text{K}$. The radial distribution of temperatures of the dust has been calculated for individual core-mantle particles. If the radial number distribution of initial interstellar dust particles is $n(r) \sim r^{-\alpha}$ then the ratio of the number with fully evaporated volatiles to the number of crystalline silicates is (assuming a spherical distribution)

$$\frac{r^{3-\alpha}(T \geq 300)}{r^{3-\alpha}(T_c)} = \frac{\beta_{iev}}{\beta_c} \quad (1)$$

where T_c is the assumed minimum crystallinity temperature; β_{iev} is the fractional number of dust grains with fully evaporated ice and β_c is the fractional number of crystallized silicates. Using $\alpha \approx 2$, $T_c=875\text{K}$, $r(T_c)=0.7\text{AU}$, $r(300\text{K})=4.2\text{AU}$ and $\beta_c = 0.15$ the amount of recondensed to initial amorphous ice is $\geq 90\%$. Since H_2O which is recondensed at any region in the presolar nebula is crystalline (Kouchi et al 1994) the comet nucleus contains at least 90% crystalline ice! Whether or not the interior of comet nuclei consists of pure amorphous ice is

not fully provable but the general evolutionary properties of comet nuclei appear to be best followed from the idea that it is *initially* amorphous (Klinger 1980; Haruyama et al 1993; Tancredi et al 1994).

In addition the volatile chemical distribution appears to be closer to the interstellar dust than to the protosolar chemical composition (Mumma et al 1993). In particular the CO/CH₄ ratio would appear to be incompatible with 90% of the chemical processes having occurred in the solar nebula.

4. Silicate metamorphosis in comet dust

Yamamoto (Yamamoto et al 1996) has proposed a novel way to produce a level of crystallinity in the silicate in moderately heated comet dust. Even if one considers that the presolar system dust has never been heated beyond about 30 K, the radicals remaining in the icy mantles which have been photoprocessed at $T \leq 15K$ will have been totally annealed away in times much, much smaller than the aggregation time scale (Greenberg et al 1993). However, any remaining radicals in the organic refractory mantles may well persist not only during aggregation but also in the billions of years since the birth of the solar system. Because of exponential reduction in diffusion rates with well depth, a relatively moderate increase in radical storage depths is all that is required. However, when the comet dust come off the nucleus its temperature, even at 2 AU from the Sun can reach temperatures of 300–500 K. This is in itself insufficient to crystallize the silicates but, if radical recombination occurs in the organic refractory mantle, radical-radical combination energy of $\sim 1 - 2$ eV per reaction can be released at the core-mantle interface. A preliminary calculation by Yamamoto et al (1996) suggests that to a depth of at least several molecules this can lead to enough local heating to produce crystallinity. Given a mean silicate radius of $0.07 \mu m$, and a mean molecule diameter of 5 \AA , a 20% fraction of crystallinity corresponds to a depth of 10 molecules. Whether the process is sufficient to penetrate to this depth is currently under investigation. If it is possible, the presence of significant crystallinity in comet dust does not require the precometary interstellar dust ever to have been heated beyond 30 K. Thus the $11.3 \mu m$ emission component of comet dust could be consistent with cold aggregation of interstellar dust.

5. $10 \mu m$ emission by comet dust

In view of the fact that only small ($\sim 1 \mu m$) size silicate particles can reproduce the observed width of the $10 \mu m$ emission of comet dust it is tempting but inconsistent to consider them as the direct source of the spectral feature. Using the scattering/absorption properties of only such small particles ignores the observed comet dust mass distribution. A consistent way of calculating the spectral energy distribution of comet dust was described by Greenberg and Hage (1990) in terms of aggregation of submicron silicate core-organic refractory mantle interstellar dust particles. We shall follow the same procedure here without discussing the details of the scattering calculation in which Mie theory was used for the spherical aggregates. We note that for high porosity aggregates the opti-

cal resonance by individual particle is diluted by the presence of a large vacuum fraction.

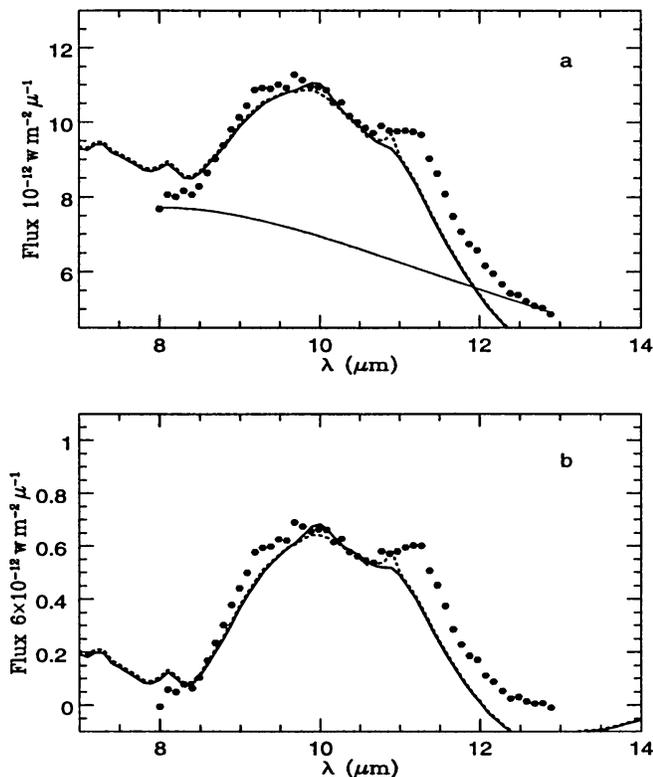


Figure 2. a) The observational and theoretical spectra of comet Halley at 0.79 AU: points — observational data (Campins & Ryan 1989); solid line — model result assuming 6% pre-aggregation crystallinity; dotted line — the same as solid line but with the $9.7 \mu\text{m}$ absorptivity of crystalline silicate reduced to $1/2$; thin solid line — the assumed dust thermal emission. b) The same as a but for the silicate excess emission (i.e., with the dust thermal emission subtracted).

We represent comet dust as spherical aggregates of core-mantle particles with uniform porosity P and size distribution as given by McDonnell et al (1989). The size distribution undoubtedly depends both on the comet solar distance and on the radial distance of the dust from the nucleus. There is abundant evidence for some fragmentation with distance. There also appear differences in the spectral emissivity of different comets with varying $11.3 \mu\text{m}$ structure (Hanner et al 1994a). However our first aim will be to determine whether, within some framework, varying mainly the degree of crystallinity may lead to some of the main features of comet dust emissivity in the $10 \mu\text{m}$ region. We have considered porosities from 0.93 to 0.975 but the following results are limited to $P = 0.95$.

5.1. Pre-aggregation crystallization

We model the dust as consisting of a mixture of pure crystalline silicates (as defined by Mukai & Koike 1990) mixed in with amorphous silicate core-organic refractory mantle particles with the mantles equal in mass to the cores. In Fig.2 the percentage of crystalline to amorphous silicate is 6%. Fig.2a shows the spectral energy distribution and Fig.2b shows the net emission at $10 \mu\text{m}$ after dust thermal continuum subtraction. As already noted earlier (Greenberg & Li 1995) the short wavelength side of the $10 \mu\text{m}$ emission is dominated by the presence of the organic refractory mantle absorptivity. Without such mantles the short wavelength side would be substantially too low compared with the comet emission. It is obvious that with only 6% crystallinity the $11.3 \mu\text{m}$ feature is not produced. In view of uncertainties in the spectral properties derived by Mukai & Koike (1990) we have tried to estimate what effect there would be if the short wavelength portion of their crystalline absorptivity were reduced, thus enhancing the relative effect of the $11.3 \mu\text{m}$ feature. This effect occurs but the combined optical properties in the ensemble (by Maxwell-Garnett (1904) effective medium theory) appear to result in a shift of the crystalline feature to significantly shorter wavelength. What prescription one requires to provide the observed peak wavelength is not clear.

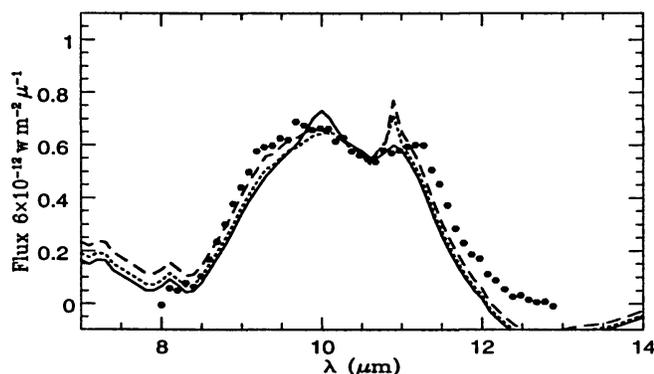


Figure 3. The observational and theoretical excess emission of comet Halley at 0.79 AU: points — observational data (Campins & Ryan 1989); solid line — model result assuming 15% pre-aggregation crystallinity; dotted line — the same as solid line but with the $9.7 \mu\text{m}$ absorptivity of crystalline silicate reduced to $1/2$; dashed line — the same as solid line but with the $9.7 \mu\text{m}$ absorptivity of crystalline silicate reduced to 0.

Since 6% crystallinity seemed inadequate we have tried the same model assuming 15% crystallinity. The series of curves shown in Fig.3 indicate what happens using not only the full Mukai & Koike crystallinity silicate structure but also using arbitrarily reduced $9.7 \mu\text{m}$ by a factor of $1/2$ or 0. The effects are rather striking in that not only is the “ $11.3 \mu\text{m}$ ” feature enhanced it is also made much sharper. This is a property produced again by using Kramers-Kronig relation on the fluffy aggregates optical properties.

In the final analysis it appears that one requires about 15% crystallinity to produce the required extra feature “strength” but we hesitate to claim an excellent match with the observation because this extra structure is both shifted in position and modified in shape. We have also considered the dust shape effect. It is worth noting that the calculated crystalline feature is shifted close to the observed peak position if one uses elongated comet dust particles.

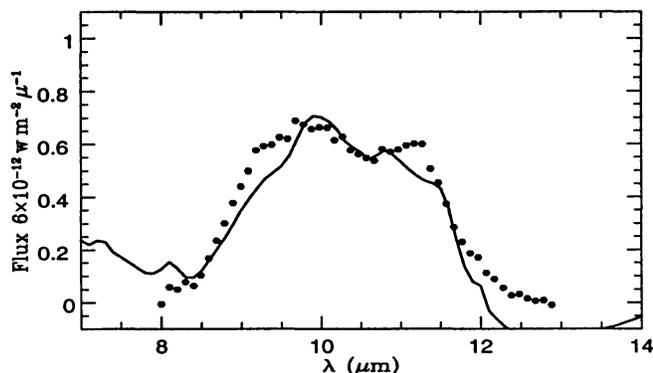


Figure 4. The theoretical excess dust emission of comet Halley by assuming 20% post-aggregation silicate crystallinity.

5.2. Post-aggregation crystallization

We have chosen to present the result only for the case where the silicate core is crystallized at its surface to 20% of its volume. The result shown in Fig.4 is not very encouraging. The strength of the crystalline feature for this morphology is largely hidden, and furthermore, even the short wavelength side of the 10 μm band appears to be reduced. We reiterate that this must be considered a very preliminary result until further examination can be made of such effects as produced by varying size distribution of the comet dust, for example.

6. Concluding remarks

We have considered two possible ways of obtaining a degree of silicate crystallinity in comet dust. The one which assumes that interstellar dust is preheated in the solar nebula leads to a model of comet dust which gives a rather good representation of the 10 μm structured emission feature with the assumption of about 15% crystallinity. However, the problem then is that the comet nucleus does not act as if it contains 85% unmodified interstellar dust but rather as if it contains less than 10% unmodified interstellar dust. If this is the case there would be substantial inconsistencies with current ideas about the nucleus consisting of predominantly amorphous ice and current interpretation of the coma material composition with respect to interstellar dust ice mantles. On the other hand, if crystallinity is a surface phenomenon on the amorphous silicate core, the dust optical properties do not provide a very good match to the observed 10 μm structure. It is our suggestion that, in spite of the lack of good agreement produced by surface crystallinity, its effects should be studied in greater depth.

Perhaps a mixture of the two processes should also be considered though it is important to bear in mind that any large degree of pre-comet silicate crystallinity implies a much larger degree of change in the comet nucleus composition and morphology.

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