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AMORPHOUS CARBON AND THE UNIDENTIFIED INFRARED BANDS

A. BORGHESI, E. BUSSOLETTI, AND L. COLANGELI
Dipartimento di Fisica, Università degli Studi di Lecce, Lecce, Italy
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ABSTRACT

We present here the absorption spectra of submicron amorphous carbon (AC) particles taken at room temperature and at three different higher temperatures in vacuum: 100°C, 260°C, and 400°C. The data show the presence of six bands which fit well the unidentified infrared emission bands (UIR) observed in space, i.e., 3.4, 3.51, 5.78, 6.29, 6.85, and 11.3 μm. The other three astronomical bands at 3.28, 7.7, and 8.6 μm are lacking. All the features are identified by means of spectroscopic analysis. The absence of the 7.7 μm band may be attributed to a CN stretching of the NH_2 functional group, which is absent in the laboratory but which may be present in sources rich in nitrogen atoms.

A comparison between the bands expected from polycyclic aromatic hydrocarbons (PAHs) and our data indicates that the former show the presence of some bands absent in AC samples while they are lacking of other features present in the AC spectrum.

This result, combined with the good agreement existing between Raman spectra of mixtures of PAHs and graphite and the Orion bar emission in the 5–10 μm region, suggests that UIR bands may be explained as produced by a collection of both amorphous carbon and PAHs coexisting together in space. This picture is particularly attractive because amorphous carbon shows also a bump at around 2350 Å quite similar to the interstellar one.

Subject headings: infrared spectra — interstellar: grains — laboratory spectra

I. INTRODUCTION

Since their discovery in the spectrum of the planetary nebula NGC 7027, the seven unidentified emission bands (UIR bands) have represented a puzzling problem in astrophysics. They occur generally in emission, respectively at 3.3, 3.4, 3.51, 6.2, 7.7, 8.6 and 11.3 μm; very often they are found together in a wide variety of galactic and extragalactic regions which are rich in UV flux. Good reviews on the subject have been reported by Aitken (1981), and recently by Allamandola (1984) and Willner (1984).

Weaker features falling at 5.6 μm and 6.95 μm have been also discovered, again in NGC 7027, by Bregman *et al.* (1983) and very recently in several planetaries, reflecting nebulae, and H II regions by Cohen *et al.* (1986). The relative intensities of the UIR bands may vary a little (Simpson *et al.* 1984) while their widths seem to be independent of the source.

The shape, width, and continuous nature of the bands are usually attributed to a solid state origin (Bregman and Rank 1975; Grasdalen and Joyce 1976; Tokunaga and Young 1980) probably due to materials that form and are destroyed together (Willner 1984). However, this idea has been recently critized by Allamandola, Tielens, and Barker (1985) and by Cohen, Tielens, and Allamandola (1985) who have suggested that the observed 30–50 cm^{-1} widths and line shapes of the narrowest bands at 3.3, 6.2, and 11.3 μm are consistent with emission from highly vibrationally excited polycyclic aromatic hydrocarbons (PAHs). The presence of the same material is suggested by Cohen, Tielens, and Allamandola (1985) to explain the plateau of emission from 11.3 to 13.0 μm detected in *IRAS* spectra of sources which show strong 7.7 and 11.3 μm features.

Actually the specific nature of materials producing the features as well as their excitation mechanism is still the subject of a wide debate (Leger and Puget 1984; Sellgren 1984; Allamandola, Tielens, and Barker 1985; Cohen *et al.* 1986).

A common indication from many authors seems, however, to suggest that "some sort" of carbonaceous materials may account for most of the observed bands (see, for example, Allamandola and Norman 1978; Allamandola, Greenberg, and Norman 1979; Duley and Williams 1981; Sakata *et al.* 1984; Allamandola 1984; Leger and Puget 1984; Allamandola, Tielens, and Barker 1985; Leger and d'Hendecourt 1985).

Over the last 2 yr we have performed (Borghesi *et al.* 1983; Borghesi, Bussoletti, and Colangeli 1985; Colangeli *et al.* 1986) an extensive laboratory survey of different kinds of submicron amorphous carbon particles covering the range 1000 Å–300 μm in order to study their morphological, physicochemical, and spectroscopic characteristics. Extinction measurements in the VUV wavelength region have shown a hump centered at about 2350 Å which shifts toward shorter wavelengths as the grain dimensions reduce. By comparing this band with the average interstellar one, the data show, however, that the agreement is still not completely satisfactory down to the minimum grain size that we have produced, 50 Å (Colangeli *et al.* 1986). Smaller particles are expected to fit better the peak wavelength and the band shape: laboratory work in this direction is presently in progress.

Particular attention has been devoted to the near-IR range observations of the samples related to the UIR bands in the last year. In this paper we present the results of our study which has been performed from room temperature up to about 400°C (heating in vacuum) which includes the temperature dependence of the bands.

The data have been compared with similar results found for other kinds of carbonaceous materials as well as with theoretical computations providing a possible, though not unique, interpretation of the actual agents which produce the UIR bands.

II. EXPERIMENTAL RESULTS

Amorphous carbon has been produced following two standard methods: (*a*) by burning hydrocarbons (benzene and xylene) in air (BE, XY) and (*b*) by striking an arc in a controled Ar atmosphere ($p = 1$ torr) between two amorphous carbon electrodes (AC). An extensive description of the experimental procedures, of the size range and morphological and structural properties of the particles has been published before (Borghesi *et al.* 1983) and will be not repeated here.

In both cases spherical grains with mean radii respectively of about 150 Å (BE,XY) and 50 Å (AC) are obtained. Observations performed by means of our electron microscope Philips EM 400 T working at 100 kV have shown, in transmission, that the image of single grains does not present any ordered structure on path lengths longer than about 10 Å. This evidence is confirmed by electron diffraction which does not evidence any structure in the diffraction pattern, confirming therefore that the particles are essentially amorphous. Obviously nothing can be said at shorter scale. On the other hand, since both kinds of particles are produced at high temperature and since this condition may lead to possible graphitization of amorphous carbon we cannot "*a priori*" exclude the existence of some ordered structure in our grains for path lengths shorter than the above mentioned limit.

A careful analysis of the IR transmission spectra has revealed that BE/XY grains show only weak bands at 6.29, 11.35, 12.05, and 13.33 μm, while the AC grains present six weak bands which match quite well the UIR bands at shorter wavelengths (see Table 1).

This difference seems to be attributed to the degree of hydrogenation of the two samples. Since the experimental conditions of the spectroscopic observations have been the same both for AC and BE/XY grains, the actual causes may be ascribed to the difference of the starting raw material and/or to a different reactivity of the grains which are formed.

In the first case, since the AC sample has been produced in an argon atmosphere, the presence of hydrogen can be considered only if it was already present in the parent electrodes.

In the second case, which actually does not exclude a previous contribution of hydrogen, we can derive that arc striking produces particles much more reactive than those obtained by burning hydrocarbons in air (see next paragraph).

The above mentioned results indicate that, among our samples, AC particles appear the most suitable to simulate carbonaceous interstellar material, and therefore we have focused our attention mainly on these particles in the following.

The amorphous carbon grains have been collected onto both KBr and NaCl substrates for spectroscopic observations while they have been deposited onto special grids to perform a parallel morphological analysis by means of transmission electron microscope (TEM) technique.

The IR transmission of each sample has been taken not only at room temperature but also, after 10 hr of stabilized heating in vacuum, respectively at 100°C, 260°C, and 400°C.

An example of the spectra is reported in Figures 1 and 2. Table 1 (cols. [2]–[5]) reports a summary of the peak wavelengths compared with those of UIR bands (col. [1]). Since, within the experimental errors, no appreciable difference is observed in the peaks at 100°C with respect to room temperature, they are not reported. Columns (6) and (7) show the observed bands detected at room temperature and at 500°C in the Quenched Carbonaceous Composite (QCC) synthesized in form of a film by Sakata *et al.* (1984). Finally, columns (8) and (9) report to expected peak wavelengths from two polycyclic

TABLE 1

INFRARED BANDS MEASURED IN THE LABORATORY FOR CARBONACEOUS MATERIAL AND THE UIR BANDS

UIR BANDS (1)	AMORPHOUS CARBON			BENZENE	QUENCHED CARBONACEOUS COMPOSITE		CORONENE	CHRYSENE	POSSIBLE INTERPRETATION OF UIR BANDS (10)
	T_{amb} (2)	260°C (3)	400°C (4)	T_{amb} (5)	T_{amb} (6)	500°C (7)	$C_{24}H_{12}$ (8)	$C_{18}H_{12}$ (9)	
3.28	...	...	...	...	3.29	...	3.3	3.3	CH stretching of $-CH$
	3.39	3.39	...	...	3.39	...	...	...	
3.4	3.42	3.42	...	...	3.42	...	...	...	Asymmetric stretching in $-CH_3$
3.51	3.51	3.51	...	...	3.48	...	...	...	Symmetric stretching in $-CH_3$
	...	...	...	...	...	...	5.2	...	
5.62	5.78	5.78	5.78	...	...	...	...	...	CO stretching in $-CHO$
6.29	6.29	6.29	6.29	6.29	6.25	6.25	6.2	6.2	Skeletal in plane C=C vibration/ $-NH_2$ deformation
	...	...	...	...	...	...	...	6.5	
6.9	6.85	6.85	...	...	6.94	...	...	6.9	Asymmetric deformation in $-CH_3$
7.27[a]	7.3	7.3	7.3	...	7.27	...	...	...	CH rocking in $-CHO$/ symmetric deformation of CH in $-CH_3$
7.7	...	...	...	...	...	...	7.6	7.8	CN stretching in $-NH_2$
	...	...	...	...	...	...	...	8.2	
8.6	...	...	...	...	...	...	8.85	8.75	In-plane aromatic CH bending
	...	...	...	...	...	...	...	10.6	
11.3	11.3	11.3	11.3	11.35	11.4	11.4	...	11.4	Out-of-plane aromatic CH bending (one adjacent H atom)
	...	...	...	12.05	11.96	...	11.9	12.2	Out-of-plane aromatic CH bending (two adjacent H atoms)
	...	...	...	13.33	13.24	...	...	...	Out-of-plane aromatic CH bending (three adjacent H atoms)

[a] Seen in NGC 7027; Bregman *et al.* 1983.

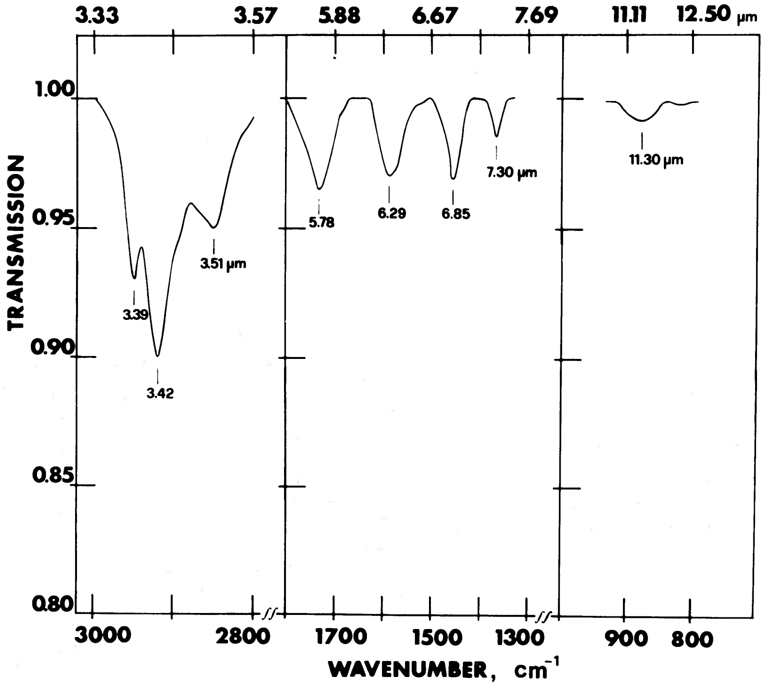

FIG. 1*a*

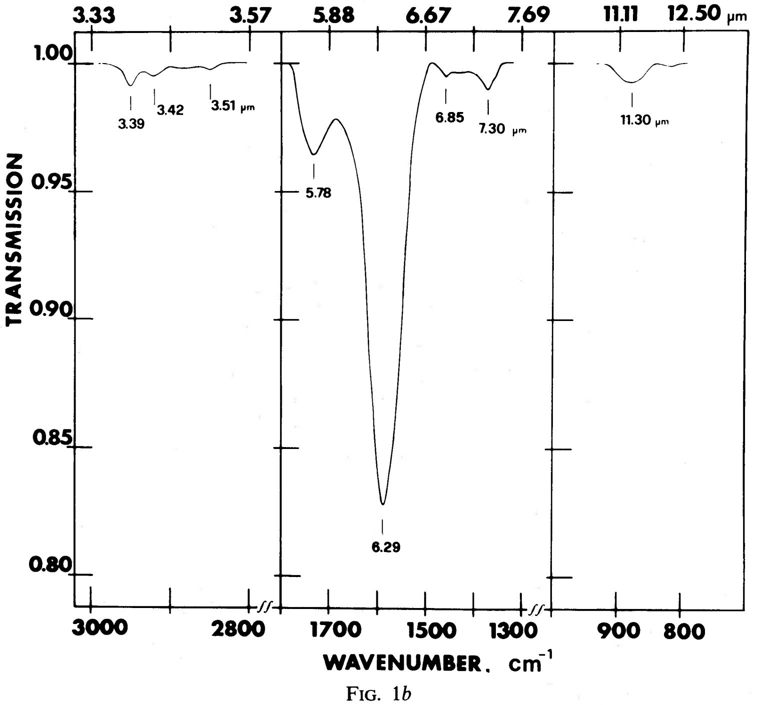

FIG. 1*b*

FIG. 1.—Transmission of amorphous carbon (AC) samples normalized to the continuum (*a*) at room temperature and after 10 hr of stabilized heating at (*b*) 260°C and (*c*) 400°C.

aromatic hydrocarbons suggested as representative of the class proposed to produce UIR bands respectively by Leger and Puget (1984) (coronene $C_{24}H_{12}$) and by Allamandola, Tielens, and Barker (1985) (chrysene $C_{18}H_{12}$).

Considering columns (1)–(4) of Table 1 we note the following:

1. Six AC bands fit well those observed in space, i.e., 3.42, 3.51, 5.78, 6.29, 6.85, and 11.3 μm.

2. Three bands are absent in our samples, i.e., 3.28, 7.7, and 8.6 μm.

By observing Figure 1 we have also noticed the following behavior when the temperature increases:

3. The bands at 3.39, 3.42, 3.51, and 6.85 μm decrease in intensity and disappear completely at 400°C according to QCC results.

4. The band at 6.29 μm increases in intensity.

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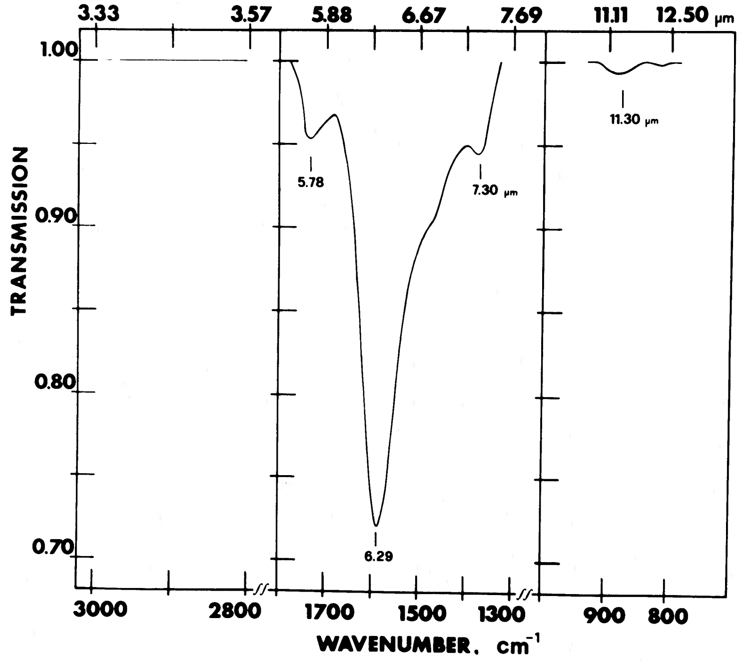

FIG. 1*c*

5. The 5.78, 7.3, and 11.3 μm bands remain stable.

Points (4) and (5) show a different behavior from that observed in QCC where all bands decrease in intensity when temperature increases, except those at 6.25 and 11.4 μm.

If all the bands disappeared in our case we should be tempted to conclude that we were facing a typical evaporation of some molecular groups adsorbed onto the surface of the grains.

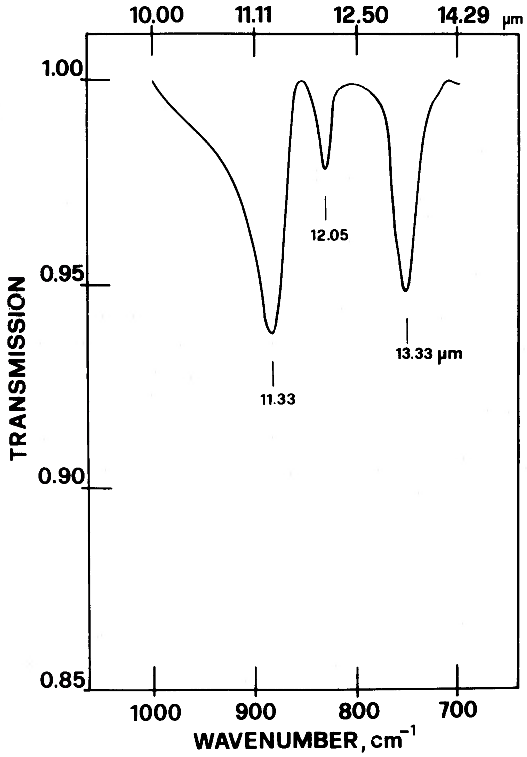

FIG. 2.—Transmission of amorphous carbon (BE) samples normalized to the continuum, at room temperature.

This is the case of the first group of bands reported in point (3) which seems to indicate a common origin, i.e., that the same functional group is the source of the different features (see next paragraph).

On the other hand, the stability of 5.78, 7.3, and 11.3 μm bands indicates that we are in presence of stronger bonds. They are essentially not affected by heating up to 400°C and may be attributed to functional groups different from those which produce group (3) bands.

Finally, the increase with temperature of the band at 6.29 μm seems to indicate again a different origin. Actually we attribute this band to skeletal in plane C=C bonds; on the other hand, heating can form C=C bonds via graphitization. Therefore, the stronger absorption observed at this wavelength in our samples may be due to the formation of an increasing number of C=C bonds in the grains as the temperature increases. It is worthwhile to note that new diffraction measurements performed on samples heated to 400°C do not show, however, patterns different from those obtained before thermal treatment. This implies that, in spite of a higher degree of graphitization produced by heating, the particles remain still essentially amorphous, within the experimental limits of our diffraction analysis.

III. DISCUSSION

The IR spectrum of amorphous carbon shows six bands whose peak wavelengths are in good agreement with six of the nine UIR bands, providing then further support to the indication that they should have a carbonaceous origin. This result is of great interest if we consider that it is the first one obtained directly from submicronic particles with dimensions close to those that are expected to be characteristic of actual interstellar grains.

According to Duley and Williams (1981) we recall that amorphous carbon is expected to be highly reactive with respect to adsorption of atoms and molecules. This is due to the fact that, in spite of the variety of forms in which amorphous carbon may exist (Boehm 1966), all are characterized by microcrystallinity produced by graphitic platelets randomly oriented in a larger grain. In this case, the interlayer distance is larger than in graphite while the C—C distance is the same; in addition, some carbon may be tetrahedrally bonded. This structure is randomly oriented with a lack of long-range order. Both effects produce a higher portion of chemically active sites where the C atoms, sitting on the periphery of crystallites, are in unsaturated states.

In space many of these reactive sites can be occupied by single H atoms or groups so that CH, CH_2, or CH_3 bonds are expected. In addition, other functional groups such as OH, CHO, or NH_2 may be present, depending on environmental conditions.

It is obvious that the shape and intensity of the astronomical bands depend critically upon the presence and the relative abundances of the different functional groups which may form in the actual local conditions. However, since H is the most abundant, we expect the various forms of CH to be dominant, in general. It is therefore not very significant, in our opinion, to compare the intensity of the UIR bands and those detected in the laboratory.

Furthermore, the question of excitation mechanisms and how these will affect the relative intensities and band profiles is still not clear. Thus, we have limited our attention to a possible identification which may help to single out the nature of the agents producing the unidentified features.

The last column in Table 1 lists the tentative interpretation that we give; it is worthwhile also to note that in some cases this is not unique as discussed in the following.

The 3.28 μm band is ascribed to CH stretching of the aromatic functional group —CH as pointed out by many authors (e.g., Duley and Williams 1981; Leger and Puget 1984; Sakata *et al.* 1984; Allamandola, Tielens, and Barker 1985).

The 3.42, 3.51, and 6.85 μm bands must be due to CH stretching and bending modes of the same functional group $-CH_3$ according to the temperature dependence observed in our sample. A similar interpretation has been already given by Duley and Williams (1981, 1983).

The laboratory spectra do not yield a band falling exactly at 5.62 μm as seen in space; the nearest one appears at 5.78 μm and may be ascribed to CO stretching of the aldehyde —CHO group. This identification is supported by the presence of the band at 7.3 μm in laboratory spectra which can be assigned again to —CHO (CH rocking) according to Duley and Williams (1981). We note that another possible cause of the band may be a symmetric deformation of CH in aliphatic $-CH_3$. We tend however to exclude this possibility on the basis of the temperature behavior of bands due to the aliphatic and aldehydic groups which are all reduced in intensity while the 7.3 μm remains stable up to 400°C. Thus, we believe it is a low-intensity aromatic band.

It is interesting to mention here that both the 5.78 μm and 7.3 μm bands have been detected in NGC 7027 (Russell, Soifer, and Willner 1977) indicating then that features other than the usual UIR bands may be present in celestial sources confirming therefore the suggestion that the existence of these bands may be strongly dependent upon specific environmental conditions.

The wavelength range between 11.0 μm and 13.3 μm is generally attributed to out-of-plane aromatic C—H bending modes where the exact position of the bands is depending on the number of adjacent H atoms present on a ring (Bellamy 1958; Duley and Williams 1981; Leger and Puget 1984). This interpretation seems to be supported from our results both on AC and BE/XY particles. AC grains are present on only the 11.3 μm band which appears, in addition, quite weak (Fig. 1*c*). To the contrary, the BE/XY spectra do show stronger bands falling at 11.35, 12.05, and 13.33 μm (Fig. 2), characteristic, respectively, of one, two, and three adjacent H atoms.

This difference indicates that the raw starting materials used to produce amorphous carbon in the laboratory still maintain, to some extent, their initial hydrogenation justifying the prevalence of rings in particles obtained by burning benzene and xylene with respect to those formed by striking an arc between two amorphous carbon electrodes.

Particular attention, even though for different reasons, must be devoted to the three bands falling respectively at 6.2, 7.7, and 8.6 μm. The first one is commonly attributed to a skeletal in-plane C=C vibration in aromatic hydrocarbons; it is present in our laboratory spectra and in theoretical emission spectra calculated for polycyclic aromatic hydrocarbons (Leger and Puget 1984; Allamandola, Tielens, and Barker 1985).

Duley and Williams (1981) report at 6.2 μm the existence of a band due to $-NH_2$ deformation of the aromatic amine group. Actually, since our ambient conditions did not contain appreciable nitrogen, the aromatic identification seems the more plausible. On the other hand we cannot exclude "*a priori*" that in space, where suitable conditions may hold, a nonnegligible contribution to the band intensity may derive from $-NH_2$ if a consistent N abundance is present. Some support to this speculation seems to come from the absence, in our data, of the 7.7 μm band which is attributed again by Duley and Williams (1981) to CN stretching of $-NH_2$; however, this assignment implies a NH stretch at 2.9 μm, a band not yet seen in astronomical spectra.

In light of the above-mentioned considerations, we tend to attribute the bands of AC samples mainly to partial hydrogenation of the grains. The heating treatment in vacuum is expected to reduce the hydrogen content so that the features due to $-CH_3$ are seen to decrease and then to disappear as temperature increases. More stable groups remain essentially unchanged at the temperatures reached in our experiment. At the same time, the band attributed to the C=C bond increases, suggesting that high temperatures tend to favor the formation of more aromatic sp^2 bond via graphitization. In this case we note, however, that diffraction measurements, within our experimental limits, do not show any measurable change of the amorphous structure of the grains. Graphitization thus is present, as indicated by the increase of the 6.29 μm band, but it seems not to alter, within our temperature range, the main particle structure.

The behavior of all the observed features seems in agreement with the above interpretations except for the persistence at higher temperatures of the weak 11.3 μm band, usually attributed to the out-of-plane aromatic —CH bending.

Actually we do not have sufficient elements to explain this discrepancy; we observe that this band is also present in the spectra reported by Sakata *et al.* (1984) for QCC after heating at ~500°C. In these conditions we suggest that other attributions may be possible. For example, the IR spectrum of graphitic clusters (Leger and Puget 1984) is characterized by very weak bands at 6.3 μm and 11.5 μm that the authors attribute to IR active lattice modes.

Since our grains are expected to have some degree of microcrystallinity, we tentatively suggest that the 11.3 μm band that we observe may be due to the joint contribution of both out-of-plane aromatic CH bending and IR lattice modes. As temperature increases, the first contribution will diminish while the latter remains stable, or even increases, in order to justify the observed persistence of the band.

A completely different interpretation of the astronomically observed band may be given if we consider PAHs as a possible source.

According to Leger and Puget (1984), Allamandola, Tielens, and Barker (1985), Cohen, Tielens, and Allamandola (1985), and Cohen *et al.* (1986), PAHs show several bands in the region around 7.7 μm due to carbon skeletal modes. We note, however, that none matches well the broad profile of the observed band and the peak wavelength (see Table 1). However, since individual peaks can vary from molecule to molecule, a collection of many PAHs may probably simulate sufficiently well the astronomical band observed at 7.7 μm. An indication in this sense comes from the results of a comparison (Allamandola, Tielens, and Barker 1985) between the Raman spectrum of a mixture of aromatic hydrocarbons and noncrystalline graphitic material. This "auto-soot" is able to reproduce quite well the shape of the 5–10 μm spectrum of the Orion bar (Bregman *et al.* 1984) with broad bands falling at 6.35 μm and 7.5 μm.

At this stage both interpretations, i.e., $-NH_2$ origin or carbon skeletal origin in PAHs, seem plausible. Specific experiment of amorphous carbon production in N-rich atmosphere are in process to better investigate the effects of N and $-NH_2$ functional groups bonded to active sites on the surface of grains.

Last is the 8.6 μm band, present in the UIR band spectrum and lacking in laboratory spectra both of amorphous carbon and QCC. Two bands are instead expected from coronene and chrysene, respectively, falling at 8.85 μm and 8.75 μm. According to Leger and Puget (1984) these bands seem to be characteristic of in-plane aromatic C—H bending which apparently is weak but active in polycyclic aromatic hydrocarbons as well as in soot acccording to the results of Allamandola, Tielens, and Barker (1985).

IV. SPECULATIONS AND CONCLUSIONS

Submicron amorphous carbon grains ($r = 50$ Å) produced by striking an arc in an Ar atmosphere show spectra with bands whose peak wavelengths fit reasonably well with six of the nine UIR bands providing further support to the idea of their carbonaceous origin. In the case of NGC 7027 one extra band at 7.3 μm is present and coincident with the feature appearing in our samples. Most of the bands are interpreted as due to functional groups attached to chemically active sites where C atoms are in unsaturated states.

Three bands at 3.28 μm, 7.7 μm, and 8.6 μm are lacking in AC grains. The absence of the 7.7 μm band in our spectra could be justified by the interpretation that it is due to $-NH_2$ (CN stretching), which may be present in space but is absent in the laboratory atmosphere. No explanation can be given for the other two bands which are respectively attributed to aromatic CH stretching and to in-plane CH bending.

UIR bands have been recently attributed to polycyclic aromatic hydrocarbons such as coronene and chrysene. The last material presents six bands fitting the peak wavelengths of UIR bands. In this case we note, however, the presence of two extra bands at about 6.5 μm and 8.2 μm and a quite strong band at 10.6 μm, comparable in intensity to that at 6.2 μm, which are not detected in celestial sources. Actually, such molecules are representative of different classes of PAHs: coronene is symmetric and shows then a simple spectrum; chrysene, on the other hand, is not, and it therefore is characterized by a more complex one.

On the other hand, spectra of PAHs do not show the 3.4, 3.5, and 5.62 μm bands, present in AC and in UIR bands, while they have a band around 7.7 μm and one at 8.75 μm absent in AC grains but detected in celestial sources.

We face therefore some sort of complementarity between the amorphous carbon spectrum which is lacking in some UIR bands and that of PAHs which covers the absent ones though extra bands which are not astronomically observed may be present.

Since the unidentified IR bands are observed in planetary nebulae, according to Allamandola, Tielens, and Barker (1985) and to Crawford, Tielens, and Allamandola (1985), it is likely that PAHs are formed in the carbon-rich outflow from these sources. A possibility exists that they may be the leftover condensation nuclei which have not been incorporated into carbon grains. We expect therefore that around these objects there may exist a mixture of both PAHs and amorphous carbon grains whose emission is able to produce the entire family of UIR bands. The solid particles should be responsible for one portion of bands while a collection of PAHs should be responsible for the remaining part. Both sources contribute evidently to the emission spectrum of common bands.

We stress that this speculation appears particularly attractive because it seems to solve many problems so far left open, and, in addition, it may be able, with the same basic material, to account also for the observed interstellar hump provided that grain dimensions smaller than 50 Å may be obtained.

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A. BORGHESI, E. BUSSOLETTI, and L. COLANGELI: Dipartimento di Fisica, Università degli Studi di Lecce, Via Arnesano, 73100 Lecce, Italy