THE INFRARED EMISSION FEATURES AND POLYCYCLIC AROMATIC HYDROCARBONS

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ABSTRACT. The infrared evidence which supports the PAH hypothesis is briefly summarized. Rather than presenting a general discussion of these assignments, this paper focuses on the spectroscopic issues raised by recent observational and experimental developments. These issues include: the position and profile of the "1310" cm⁻¹ ("7.7" μ m) feature, the position and intensities of the bands in the 910-710 cm⁻¹ (11-14 μ m) region, the newly detected 1900 cm⁻¹ (5.3 μ m) band, and the spatial and spectral variations in the 3000 cm⁻¹ (3 μ m) region as well as in the 12 and 25 μ m IRAS bands. It is concluded that the infrared evidence for interstellar PAHs and PAH-related species is compelling.

1. INTRODUCTION

The scientific organizing committee has allocated one entire day of this 5 day symposium to the interstellar emission bands at 3050, 1610, "1310," 1160 and 885 cm^{-1} (3.28, 6.2, "7.7," 8.6 and 11.3 microns) because the bands provide very detailed information about an apparently ubiquitous and abundant "dust" component. The bands are associated with a large number of different types of astronomical object (Bregman, 1989; Roche, 1989; and Sellgren, 1989; and references therein) implying that they are due to a surprisingly widespread, extremely stable, interstellar constituent. While it has been recognized for some time now that the emission was most likely due to IR fluorescence from molecule-sized species pumped by UV photons (Allamandola, Greenberg and Norman, 1979; Sellgren, 1984), the identity of the carriers has remained enigmatic. Current thinking that polycyclic aromatic hydrocarbons (PAHs) and related materials are the long sought after carriers can be traced to the suggestion of Duley and Williams (1981) that they arise from the vibrations of chemical subgroups attached to the aromatic moieties at the surfaces of small (<0.01 μ m), amorphous carbon particles. Subsequently Léger and Puget (1984) and Allamandola, Tielens and Barker (1985) (hereafter LP and ATB respectively) proposed that individual molecule-sized (>0.001 μ m) PAHs and PAH related materials were the carriers. The principal reason for this assignment was the resemblance of the interstellar emission spectra with the vibrational spectra of these materials. Related observations support an aromatic carrier as well. The fraction of total IR luminosity radiated in the "1310" cm⁻¹ ("7.7" μ m) feature, which is by far the most intense of the bands, is strongly correlated with the carbon available

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FWHH ASSIGNMENT² λ ν (cm^{-1}) (cm^{-1}) (Microns) THE MAJOR BANDS 3040 3.29 30 Aromatic C - H stretch ($v=1 \rightarrow v=0$) 30 Aromatic C - C stretch 6.2 1615 1315-1250 7.6-8.0 70-200 Blending of several strong aromatic C - C stretching bands Aromatic C - H in-plane bend 1150 8.7 885 11.3 30 Aromatic C - H out-of-plane bend for non-adjacent, peripheral H atoms THE MINOR FEATURES 3085 3.24 Overtone and/or combination involving fundamentals in the 1810-1050 cm⁻¹ (5.52-9.52 μ m) range 2995 3.34 Overtone and/or combination involving fundamentals in the 1810-1050 cm⁻¹ (5.52-9.52 μ m) range "20" Aromatic CH stretch $(v=2 \rightarrow v=1)$ 2940 3.4 2890 3.46 Overtone/combination band involving fundamentals in the 1810-1050 cm⁻¹ (5.52-9.52 μ m) range, aromatic CH stretch (high v), aliphatic CH stretch,? 2850 3.51 Aromatic CH stretch ($v=3 \rightarrow v=2$), aliphatic CH stretch, overtone/combination band involving fundamentals in the 1810-1050 cm⁻¹ (5.52-9.52 μ m) range 2810 Aromatic CH stretch (high v), aldehydic CH stretch, 3.56 overtone/combination band involving fundamentals in the 1810-1050 cm⁻¹ (5.52-9.52 μ m) range Combination of CH out-of-plane and in-plane bend, ? 5.1-5.3 30 1960-1890 1785-1755 5.6-5.7 40 Overtone of 885 cm⁻¹ (11.3 μ m) band; Aromatic C – C stretch; Carbonyl C = 0 stretch, ? 1470-1450 6.8-6.9 30 Aromatic C - C stretch, aliphatic CH deformation C - H out-of-plane bend for doubly adjacent H atoms 840 11.9 790 12.7 C - H out-of-plane bend for triply adjacent H atoms THE BROAD COMPONENTS 2940 3.5 Overlap of C - H stretching modes, shifted by anharmonic effects, 3115-2740 3.21-3.65 **"300"** with overtones and combinations of C - C stretch fundamentals in the 1670-1250 cm⁻¹ (6-8 μ m) region, aliphatic CH stretch?,? ~ 1200 ~ 8.5 **"400"** Blending of many weak aromatic C - C stretching bands 1810-1050[†] 5.52-9.52[†] 880 "160" 12 Overlap of many aromatic C - H out-of-plane bending 950-740[†] 10.5-13.5 modes for non-adjacent as well as doubly and triply adjacent peripheral H-atoms Red-Near IR Continuum Electronic transitions between low-lying levels in ionized and complexed PAHs and amorphous carbon particles Mid-IR Continuum Quasi-continuum formed by overlapping overtone and combination bands

TABLE 1: EMISSION COMPONENTS: PROPERTIES AND ASSIGNMENTS¹

" " Value estimated from several published spectra. [†] Rough limits of the feature.

1: When the assignment is not clear, several possible explanations are listed. The first seems most likely.

2: This table is extensively discussed in Allamandola, Tielens, and Barker, 1989.

in planetary nebulae (Cohen *et al.*, 1986). As the carriers must be produced under harsh conditions in planetary nebulae, they must be extremely stable. Furthermore, although there is some variation among the relative band intensities, the bands are correlated implying that a single class of chemical species is responsible (Cohen *et al.*, 1986). These observations are completely consistent with PAHs which are a family of extremely stable, planar hydrocarbons which have a chicken wire-like structure.

Interstellar PAHs are thought to be very abundant, more so in fact than all of the known interstellar, gaseous, polyatomic molecules combined (LP; ATB). The proposed ubiquity and high abundance of interstellar PAHs implies that their influence on various processes and phenomena can be profound (Omont, 1986). For example they may be responsible for the infrared cirrus discovered by the IRAS satellite (ATB; Puget, Léger and Boulanger, 1985), some of the diffuse interstellar bands in the visible (van der Zwet and Allamandola, 1985; d'Hendecourt and Léger, 1985; Crawford, Tielens, and Allamandola, 1985), maintaining interstellar cloud temperatures (d'Hendecourt and Léger, 1987), moderating interstellar cloud chemistry (Lepp and Dalgarno, 1987), and contributing to the deuterium enrichments found in interplanetary dust particles and meteorites (Allamandola, Sandford and Wopenka, 1987; Allamandola, Tielens and Barker, 1989b).

In order to ascertain just how seriously one should take these suggestions it is important to consider the spectroscopic arguments in favor of PAHs. While definitive evidence is lacking in the ultraviolet and visible spectral regions (Donn, Allen, and Khanna, 1989; Léger *et al.*, 1989), the case in the infrared is now compelling. Several predictions have been borne out and the theory continues to be capable of accommodating new observational results. Some of the recent additions to the evidence that PAHs and PAH-related species are responsible for many aspects of the infrared emission spectrum will be reviewed in this paper.

2. INTERSTELLAR SPECTRA AND THE SPECTRA OF PAHS

The interstellar emission spectra consist of strong, well-defined bands, weak bands, and broad features. To date, the relative intensities of the strong bands appear to be reasonably well-correlated. This is not the case for the weak bands and broad features. Any model which attempts to explain the IR emission phenomenon should address all of these characteristics. With one exception, the proposals skirt most of the issues of spectroscopic detail. There are several interpretations of the emission phenomenon based on PAHs. Duley and Williams (1988, 1989), while not specifically addressing the weak bands and broad features, envision that the well-defined bands arise from PAHs and side-groups on the PAHs which comprise hydrogenated amorphous carbon (HAC) particles. Léger and d'Hendecourt (1987, 1989), again principally addressing the well-defined bands, envision that they arise from free, molecule-sized PAHs containing between 50-100 carbon atoms and not PAHs which are part of carbonaceous particles. Allamandola, Tielens and Barker (1987, and this paper) envision that smaller, free, ionized PAHs, containing between 20-50 carbon atoms, are responsible for both the well defined and weak bands but larger PAHs, PAH clusters, and/or amorphous carbon particles, containing on the order of 400 carbon atoms, contribute to the broad features. Whether the PAHs are free or part of a larger particle is, at present, controversial. For the emission to originate from the aromatic structural units of a particle (HAC), or PAH cluster,

requires that the vibrational energy created by the absorption of the exciting photon remain localized in a specific PAH unit long enough for it to radiatively relax in the IR ($\approx 10^{-2}$ s). The principle objection to this is that the timescale for normal nonradiative relaxation in a solid is usually many orders of magnitude faster. While one might imagine that some individual PAH units may be very loosely coupled to the remainder of the carbon particle, this must be demonstrated experimentally before it can be accepted. On the other hand, IR emission from highly vibrationally excited, free molecules is expected. In fact, the detection of UV-pumped IR emission from the bicyclic aromatic molecule azulene has been reported by Cherchneff and Barker (1989).

In view of the space limitation, this article will not deal with the differences between the various models but rather focus on the underlying, unifying theme that PAHs and PAH-like species are responsible for the emission. Furthermore, rather than discuss the general assignments yet again, this paper will focus on the spectroscopic issues raised by recent observational and experimental developments. All of the IR emission features and assignments are summarized in Table 1. The emission spectrum from position 4 in the Orion Bar is compared with the absorption spectra of several PAHs in Figure 1. Although the interstellar spectra resemble the laboratory data, they do not match in detail. Some questions raised by a careful comparison of the interstellar spectra with a random set of laboratory spectra of PAHs are:

- 1. Why does the "1310" cm⁻¹ ("7.7" μ m) feature seem so poorly reproduced?
- 2. Why do so few of the bands in the 910-710 cm⁻¹ (11-14 μ m) range in the laboratory spectra match the single interstellar band at 885 cm⁻¹ (11.3 μ m) and why are the laboratory features far more intense with respect to the bands at higher frequencies than in the interstellar case?
- 3. All of the laboratory spectra show a weak to moderate band near 1900 cm⁻¹ (5.2-5.3 μ m), is there a corresponding interstellar feature?

In addition, recent observations show specific details which are not immediately apparent from the laboratory data. The questions raised here are:

- 1. Can PAHs account for both the spectral and spatial variations found in the 3230-2750 cm⁻¹ (3.1-3.6 μ m) region?
- 2. Can PAHs account for the spatial variation in the IRAS 12 and 25 μ m bands in reflection nebulae and the IR cirrus?

Each of these will be discussed in turn. It will be shown that the properties of PAHs permit one to satisfactorily answer all of these questions.

2.1. THE "1310" cm^{-1} ("7.7" μm) BAND

The principal difficulty here is to select a mixture of PAHs which can reproduce the position and profile of the interstellar feature. In the interstellar medium less stable PAHs will be weeded out, leaving a mixture dominated by the most stable structures. The most stable PAHs are symmetric and in the most condensed form



Fig. 1. The 3-13 micron emission spectrum from the Orion Bar compared with the absorption spectra of the PAHs chrysene, pyrene and coronene suspended in *KBr* pellets. (Orion, Bregman et al., 1988; Chrysene, Cyvin et al., 1982a; Pyrene, Cyvin et al., 1979; Coronene, Bakke et al., 1979; Cyvin et al., 1982b). Figure reproduced from Allamandola, Tielens and Barker, 1987.

(van der Zwet and Allamandola, 1985; Crawford, Tielens, and Allamandola, 1985). One example of a mixture comprised largely of the most stable molecular forms is the soot formed in the high temperature combustion of hydrocarbons. Figure 2 shows a comparison of the IR emission spectrum in the 1670-1250 cm⁻¹ (6-8 μ m) region from Orion with the Raman spectrum of auto soot.

The Raman spectrum of soot principally probes the aromatic carbon-carbon stretching vibrational frequencies because the Raman scattering cross section for these bonds is very large and they are the most dominant type of bond in the mixture. This comparison dramatically shows that the C-C vibrational frequencies in a mixture of the more stable PAHs coincides quite well with the frequency range of the interstellar features (ATB). The confident assignment of this interstellar feature primarily to symmetric interstellar PAHs is supported by the good match of the major emission bands in the 2000 to 1000 cm⁻¹ (5-10 μ m) region from NGC 2023 (dots) with the <u>calculated</u> infrared emission spectra from 4 symmetric PAHs shown in Figure 3 (Léger and d'Hendecourt, 1987).



Fig. 2. Comparison of the 5 to 10 micron Raman spectrum of auto soot (a form of amorphous carbon which is rich in the more stable PAHs) with the emission from Orion (soot spectrum, adapted from Rosen and Novakov, 1978; Orion, Bregman *et al.*, 1988). Figure reproduced from Allamandola, Tielens and Barker, 1985.

2.2. THE 900-500 cm^{-1} (11-20 μm) REGION

2.2.1. The 900-500 cm⁻¹ (11-20 μ m) band frequencies

The spectra of PAHs studied in the laboratory have several bands in this region because they have more than one H atom per edge ring. These bands have long been used by chemists as a diagnostic of the number of adjacent hydrogen atoms on the edge rings of aromatic molecules. One difficulty in assigning the emission in this region to PAHs arose because most interstellar spectra appeared to have only one band in this region at a position (885 cm⁻¹, 11.3 μ m) which implied one hydrogen per ring. This was taken to indicate severe dehydrogenation in spite of the fact that hydrogen is greater than 10⁷ times more abundant than the proposed PAHs.

This problem was resolved when it was shown that IRAS spectra of objects emitting the bands also have a broad plateau of emission from about 950-770 cm⁻¹ (10.5-13 μ m) (Cohen, Tielens, and Allamandola, 1985). This relieved the difficulties associated with invoking partial dehydrogenation. This also implied that the interstellar PAHs responsible had edge rings with single, as well as doubly and triply adjacent H atoms, but not four or five, placing strong constraints on the molecular structures possible and again indicating that the most stable, symmetric forms are dominant in the interstellar medium.



Fig. 3. Emission spectra of several compact PAHs <u>calculated</u> from laboratory absorption spectra measured at room temperature compared to the observed spectrum of the reflection nebula NGC 2023 (dots). Figure reproduced from Léger and d'Hendecourt, 1987.

Recent observations by Witteborn *et al.* (1989) and Roche, Aitken, and Smith (1989) have shown that there is resolvable structure on this plateau. The new features are just where one would expect to find them for PAHs with doubly and triply adjacent H atoms per edge-ring: in the 840-830 cm⁻¹ (11.9-12 μ m) and 790-775 cm⁻¹ (12.6-12.9 μ m) regions.

2.2.2. The 900-500 cm^{-1} (11-20 μm) band intensities

The interstellar 950-770 cm⁻¹ (10.5-13 μ m) plateau and 885 cm⁻¹ (11.3 μ m) band are generally much less intense than the "1310" cm⁻¹ ("7.7" μ m) feature whereas the opposite is true in the laboratory spectra. This discrepancy has been shown to be the result of a spectral artifact produced by the techniques used to measure most of the PAH spectra published to date. Most laboratory spectra are obtained by mixing PAH crystallites with salt and pressing the mixture into a transparent pellet. Individual PAH molecules are not isolated in this way and molecular interactions between adjacent PAHs produce large band strength enhancements in the 900-500 cm⁻¹ (11-20 μ m) region. (These experiments, reported by Sandford and Allamandola at the Wyoming Summer School on Interstellar Processes held in July, 1986, are described in detail in Allamandola and Sandford, 1988).

This property is not only important because it reconciles an apparently serious inconsistency with the PAH hypothesis but also because it provides a powerful spectroscopic argument in favor of the well-defined bands arising from free, molecule-sized species rather than particles (in which interactions would be expected to enhance emission in this region).

2.3. THE 1900 cm^{-1} (5.2-5.3 μm) FEATURE

As most laboratory spectra of PAHs show a weak band close to this position, the PAH hypotheses predicts an interstellar band here. This was recently searched for, and found in the spectrum of $BD+30^{\circ}3639$, an object which emits the well-defined bands (see Bregman, 1989; and Allamandola *et al.*, 1989*b*).

2.4. THE SPECTRAL AND SPATIAL VARIATIONS IN THE 3230-2750 cm⁻¹ (3.1-3.6 μ m) REGION

In addition to the prominent 3050 cm^{-1} (3.28 μ m) band and the broad, weak component from $3225-2780 \text{ cm}^{-1}$ (3.1-3.6 μ m), several weak features have been discovered in this region as well (Geballe *et al.*, 1985; de Muizon *et al.*, 1986). The weak features are superimposed on the broad component and can arise from several types of vibrations associated with PAHs. These include: combinations and overtones involving *CC* stretching fundamentals in the 1700-1100 cm⁻¹ (5.9-9 μ m) range, $v = 3 \rightarrow 2$ and $2 \rightarrow 1$ aromatic *CH* stretch transitions in the most highly vibrationally excited PAHs, and $v = 1 \rightarrow 0$ aliphatic *CH* stretch transitions for aliphatic side-groups attached to PAHs (de Muizon *et al.*, 1986; Barker, Allamandola, and Tielens, 1987).

The relative intensities of all of these features have recently been found to vary as a function of position near the Orion Bar and near the star exciting the Red Rectangle, indicating that several components are responsible (Geballe *et al.*, 1989). These observations, which can be accommodated by the PAH hypothesis in a straightforward way, yield insight into the size and photochemical evolution (dehydrogenation) of interstellar PAHs. The spectral and spatial variations imply that overtones and combinations of *CC* stretching bands and aromatic $CH v = 3 \rightarrow 2$ and $2 \rightarrow 1$ transitions in PAHs with 20-50 carbon atoms contribute to the broad component and weaker features in both objects. Aliphatic *CH* stretching vibrations in side groups can also contribute in the Orion Bar but are difficult to reconcile with the observed variations in the Red Rectangle (Geballe *et al.*, 1989).

2.5. THE SPATIAL VARIATION IN THE 12 AND 25 µm IRAS BAND INTEN-SITIES IN REFLECTION NEBULAE AND THE IR CIRRUS

The intensity of the 12 μ m IRAS band drops with respect to that of the 25 μ m band in the vicinity of the exciting star in the case of reflection nebulae (Sellgren, 1989; and Luan *et al.*, 1989) and close to hot stars in the case of the cirrus (Puget, 1989; and Boulanger *et al.*, 1988). The relative intensities are also found to vary between



Fig. 4. A schematic composite absorption spectrum for the out-of-plane vibrations in the mixture of aromatic molecules naphthalene, chrysene, pyrene and coronene in the 1000-130 cm⁻¹ (10-75 μ m) range compared to the IRAS bands. The regions in which the *CH* in- and out-of-plane and carbon plane bending vibrations fall are also indicated. Figure adapted from Allamandola, Tielens and Barker, 1987.

clouds of IR "cirrus" (Verter, Magnagni, and Dwek, 1989). If PAHs and PAHrelated materials are responsible for the emission, this behavior can be understood in terms of dehydrogenation rather than grain destruction. As shown in Figure 4, the 12 μ m IRAS band covers the 1250-670 cm⁻¹ (8-15 μ m) range which primarily encompasses the CH in-plane and out-of-plane bending vibrations in PAHs while the 25 μ m IRAS band covers the 560-330 cm⁻¹ (18-30 μ m) region which overlaps with many of the C - C - C aromatic, plane bending vibrations of PAHs (Allamandola, Tielens and Barker, 1987). Consequently, if a PAH is stripped of hydrogen it will lose most of its IR activity in the 1000-500 cm⁻¹ (10-20 μ m) region (IRAS 12 μ m band) but not in the 500-10 cm⁻¹ (20-1000 μ m) region. Small PAHs (15-30 carbon atoms) contribute strongly to the infrared emission because they are the most highly vibrationally excited by the absorption of an ultraviolet photon. These are also the PAHs which will be the first to suffer H atom loss if sufficiently excited (Tielens et al., 1987; Allamandola, Tielens, and Barker, 1989b). Consequently, if PAHs are indeed responsible for this emission, the drop in intensity of the 12 μ m IRAS band with respect to the 25 μ m IRAS band can be understood in terms of the dehydrogenation of the most intensely emitting PAHs and not the destruction of the carbon skeleton or of a particular class of grains. By inverting the dehydrogenation treatment described in Geballe et al. (1989), one can probe the strength

of the interstellar radiation fields in these regions.

This explanation can be tested, provided the spatial behavior of the 3050 cm^{-1} (3.28 μ m) band can be measured. As this band is due to the aromatic CH stretch, it too should show a similar drop in intensity with respect to the predominantly CCmodes in the 1670-1180 cm⁻¹ (6-8.5 μ m) and 500-10 cm⁻¹ (20-1000 μ m) regions.

3. CONCLUSIONS

Most of the spectroscopic objections to PAHs carrying the infrared emission bands can be satisfactorily answered. The general arguments in favor of PAHs and PAHrelated materials being responsible for most, if not all, of the spectral features are compelling (Table 3). Nonetheless some important questions have recently been raised. For example, what is the reason that the width of the 3050 cm^{-1} (3.28 μ m) band may vary significantly (Tokunaga et al., 1988), do QCC's contain PAHs (Sakata, 1989), do QCC's and amorphous carbon particles show the fine spectroscopic details and variations that free PAHs do (Bussoletti and Colangeli, 1989) and are the PAH structural units in amorphous carbon particles indeed loosely enough bound to permit them to radiate as free molecules (Duley, 1989)? As with most of the previous questions (which were originally perceived to be condemning to the PAH hypothesis), the answers to these should provide further insight into the nature of interstellar PAHs and the environments from which they emit.

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