

MODELING THE UNIDENTIFIED INFRARED EMISSION WITH COMBINATIONS OF POLYCYCLIC AROMATIC HYDROCARBONS

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ABSTRACT

The infrared emission band spectrum associated with many different interstellar objects can be modeled successfully by using combined laboratory spectra of neutral and positively charged polycyclic aromatic hydrocarbons (PAHs). These model spectra, shown here for the first time, alleviate the principal spectroscopic criticisms previously leveled at the PAH hypothesis and demonstrate that mixtures of free molecular PAHs can indeed account for the overall appearance of the widespread interstellar infrared emission spectrum. Furthermore, these models give us insight into the structures, stabilities, abundances, and ionization balance of the interstellar PAH population. These, in turn, reflect conditions in the emission zones and shed light on the microscopic processes involved in the carbon nucleation, growth, and evolution in circumstellar shells and the interstellar medium.

Subject headings: infrared: ISM: lines and bands — ISM: individual (Orion Bar, IRAS 22272+5435) — line: formation — line: identification — line: profiles — molecular data — radiation mechanisms: nonthermal

1. INTRODUCTION

The most prominent of the widespread interstellar, unidentified infrared (UIR) bands are the narrow (30 cm^{-1} FWHM) features at 3050 , 1610 , and 890 cm^{-1} (3.28 , 6.2 , and $11.2\text{ }\mu\text{m}$) and the strong emission envelope centered near 1300 cm^{-1} ($7.6\text{ }\mu\text{m}$), which shows a distinct shoulder at 1165 cm^{-1} ($8.6\text{ }\mu\text{m}$; Russell, Soifer, & Willner 1977; Aitken 1981; Willner 1984; Phillips, Aitken, & Roche 1984). The emission spectra also typically exhibit several substantially weaker features and underlying continua. While early observations left the impression that the spectra were almost indistinguishable from one object to another, important differences in spectral detail have emerged over time (Geballe et al. 1992; Roelfsma et al. 1996; Laureijs et al. 1996; Onaka et al. 1996; Matilla et al. 1996; Sloan et al. 1997). Given the prevalence of this emission across all phases of the interstellar medium (ISM), understanding its source has become one of the key problems in galactic astrophysics.

The idea that mixtures of UV-excited, gas-phase, polycyclic aromatic hydrocarbons (PAHs) are the band carriers was put forward over a decade ago (Leger & Puget 1984; Allamandola, Tielens, & Barker 1985, hereafter ATB). As a class, PAHs have IR spectra that are consistent with characteristic frequencies of the interstellar bands. All other classes of organics (i.e., aliphatic hydrocarbons-alkanes, alkenes, and alkynes, including carbon chains, as well as chemical subgroups such as methyl, ethyl, amino, carbonyl, carboxyl, cyano, etc.) and inorganics exhibit distinctive, characteristic bands at positions where only weak or no interstellar features are observed, and thus they represent no more than a tiny fraction of the emitting material in the emission regions. Nevertheless, since the emission arises not from a single species but from a complex *mixture* of PAHs, and since all PAHs are constructed from the same basic benzenoid structural unit, the interstellar spectra reflect an unresolvable overlapping of a myriad of individual, approximately 30 cm^{-1} wide, molecular features. While this implies that a specific molecular PAH identification is unlikely to be based on mid-IR spectroscopy alone, as shown below the interstellar infrared spectrum still provides a powerful probe of the band carriers and the ISM.

Initially, the principal case for interstellar PAHs was the

resemblance of the interstellar *emission* spectra to the then available infrared *absorption* spectra of neutral PAHs in organic solvents or crystals, but the laboratory database available at that time was not adequate for a critical evaluation of the PAH hypothesis since the conditions in solvents and crystals strongly perturb the PAH spectra and are far from those in the ISM. Thus, while the interstellar band frequencies were highly suggestive of PAHs, there were alarming relative intensity discrepancies between the interstellar spectra and the then available laboratory spectra.

In recent years, we have seen a renaissance in PAH studies that are specifically motivated to address astrophysical questions. Groundbreaking experimental work at a number of institutions has led to the development of special laboratory techniques for determining the intrinsic infrared properties of PAH molecules and PAH ions under astrophysically relevant conditions as well as PAH stabilities and reactivities (Cherchneff & Barker 1989; Shan, Suto, & Lee 1991; Brenner & Barker 1992; Schlemmer et al. 1994; Williams & Leone 1995; Szczeplanski, Wehlburg, & Vala 1995 and references therein; Hudgins & Sandford 1998c and references therein; Jochims, Baumgartel, & Leach 1998 and references therein; LePage et al. 1998; Cook & Saykally 1998). Similarly important breakthroughs have been made in the application of high-level *ab initio* methods for determining the intrinsic infrared properties of isolated PAH molecules and PAH ions (DeFrees et al. 1993; Langhoff 1996; Bauschlicher & Langhoff 1997 and references therein).

This Letter focuses on the database of individual PAH spectra generated in the Astrochemistry Laboratory at NASA Ames and presents the first application of this database to the interstellar infrared emission bands. The matrix isolation technique has been employed to study isolated neutral and ionized PAHs in a cold, essentially nonperturbing environment. At present, this is the only means by which the infrared spectra of ionized PAHs can be measured under conditions that approach those of the ISM. In this technique, a particular PAH vapor is co-condensed with a great overabundance of argon gas onto a cold (10 K), infrared-transparent window, resulting in a quasi-gas-phase condition that reproduces the low-rotational temperatures of the interstellar emitters. Under these conditions,














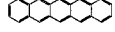






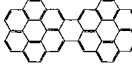


Formula	Name	Structure	Ionization State	Formula	Name	Structure	Ionization State
C ₁₀ H ₈	Naphthalene		0,+	C ₂₀ H ₁₂	Benzo(j)fluoranthene		0,+
C ₁₂ H ₈	Acenaphthylene		0		Benzo(k)fluoranthene		0,+
C ₁₄ H ₁₀	Anthracene		0,+		Benzo(e)pyrene		0,+
	Phenanthrene		0,+		Perylene		0
C ₁₆ H ₁₀	Fluoranthene		0,+	C ₂₀ H ₁₄	9,10-Dihydrobenzo(e)pyrene		0,+
	Pyrene		0,+	C ₂₂ H ₁₂	Benzo[ghi]perylene		0,+
C ₁₈ H ₁₀	Benzo[ghi]fluoranthene		0	C ₂₂ H ₁₄	Pentacene		-,0,+
C ₁₈ H ₁₂	1,2-Benzanthracene		0,+	C ₂₄ H ₁₂	Coronene		0,+
	Chrysene		0,+		Hexabenzocoronene-A		0,+
	Tetracene		0,+	C ₄₂ H ₁₈			
	Triphenylene		0	C ₄₈ H ₂₀	Dicoronylene		0,+
C ₂₀ H ₁₂	Benzo(a)fluoranthene		-,0,+				
	Benzo(b)fluoranthene		0,+				

FIG. 1.—The inventory of PAHs included in the infrared spectral database

infrared band positions differ from their corresponding gas-phase values by significantly less than 1% (typically between 5 and 10 cm⁻¹).

To date, we have measured the infrared spectra of the 23 neutral and 19 cationic PAHs listed in Figure 1 (Hudgins, Sandford, & Allamandola 1994; Hudgins & Allamandola 1995a, 1995b, 1997; Hudgins & Sandford 1998a, 1998b, 1998c). Among other species, the data set includes all of the thermo-

dynamically most stable PAHs through coronene (C₂₄H₁₂), i.e., the species most likely to be the most abundant of the smallest interstellar PAHs; dicoronylene (C₄₈H₂₀), one of the largest molecules ever isolated in a matrix; and the simplest members of the fluoranthene family, PAHs containing pentagonal rings, which are the species predicted to be important in the early stages of circumstellar PAH formation (Keller 1987). Detailed discussions and applications of the database to the astrophysical problem will be presented elsewhere (Hudgins et al. 1998), and so will a more comprehensive astrophysical model in which PAH *emission* spectra are calculated as a function of the local radiation field, charge balance, extinction, density, etc. (Bakes et al. 1998).

One of the important results of the laboratory and theoretical studies on neutral and ionized PAHs is the remarkably dramatic effect ionization has on their infrared spectra, as illustrated in Figure 2 (see, e.g., DeFrees & Miller 1989; Szczepanski et al. 1995 and references therein; Hudgins & Sandford 1998c and references therein; Bauschlicher & Langhoff 1997 and references therein). Ionization alters the IR spectrum to such an extent that the relative intensity mismatches between the interstellar bands and the laboratory spectra of neutral PAHs are resolved qualitatively, alleviating one of the more troubling *qualitative* difficulties of the interstellar PAH model.

2. MODELING INTERSTELLAR SPECTRA WITH PAH MIXTURE SPECTRA

Within the framework of the PAH model, the interstellar spectrum arises from the combined emission of a complex *mixture* of PAHs. Previous comparisons between the interstellar features and laboratory spectra have involved only individual PAHs or combinations of a very few PAHs. The large database of cold, isolated, ionized PAH spectra now alleviates this shortcoming and, furthermore, demonstrates that these data *quantitatively* resolve many of the key spectroscopic issues asso-

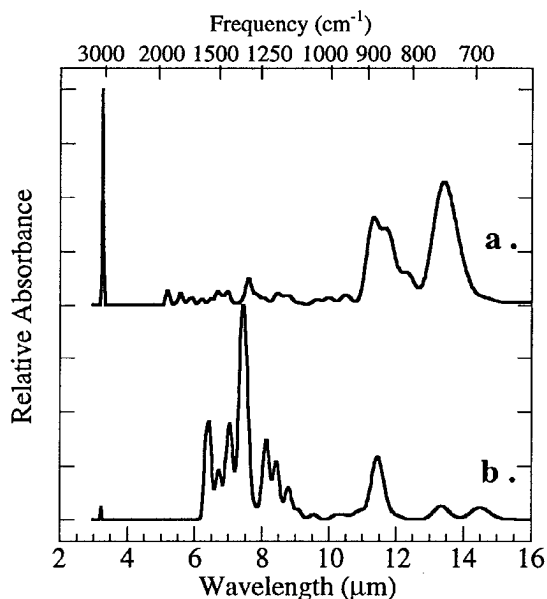


FIG. 2.—The absorption spectrum produced by the co-addition of (a), six neutral PAH spectra compared with the spectrum produced by the same PAHs in (b), the cationic form (anthracene, tetracene, 1, 2-benzanthracene, chrysene, pyrene, and coronene; see Fig. 1). Note the extreme effect that the ionization state has on relative intensities compared with its rather minor influence on peak frequencies.

ciated with frequency matching and relative band strengths. Figure 3 compares the *emission* spectra from two astronomical objects that sample very different stages in the life cycle of interstellar carbon with the lab *absorption* spectra produced by co-adding different PAH spectra. Each absorption line has been assigned a 30 cm^{-1} FWHM Gaussian profile, consistent with the natural line width expected from each emitting molecule (ATB; Allamandola, Tielens, & Barker 1989, hereafter ATB2). This natural line width arises from intramolecular vibrational energy redistribution; it does *not* arise from the blending of individual rotational lines in the emitting molecules that remain *rotationally* cool throughout the excitation/emission process (see, e.g., Brenner & Barker 1992; Williams & Leone 1995; Cook & Saykally 1998). The spectra in our database are then scaled and co-added until good agreement with the astronomical spectrum is achieved. The co-added spectra *do not* include the $5\text{--}10 \text{ cm}^{-1}$ matrix shift or the redshift expected between the absorption peak frequencies and the emission frequencies of vibrationally excited PAHs (Flickinger, Wdowiak, & Gomez 1991; Brenner & Barker 1992; Colangeli, Mennella, & Bussoletti 1992; Williams & Leone 1995; Joblin et al. 1995; Cook & Saykally 1998) since these effects are less than the natural line width.

Two cases are given that illustrate the ability of the PAH infrared database to accommodate both the overall appearance *as well as* the spectral variety exhibited by the different classes of astronomical objects that produce the IR emission features. The proto-planetary nebula phase likely represents the earliest stage in the life cycle of cosmic PAHs (ATB; ATB2; Gauger, Gail, & Sedlmayr 1990; Cherchneff, Barker, & Tielens 1992; Cadwell et al. 1994). Figure 3 compares the spectrum from one such object, IRAS 22272+5435, with the spectrum of a PAH mixture generated from our spectral database. IRAS 22272+5435 is a carbon-rich object transiting from an asymptotic giant branch (AGB) star to a planetary nebula (Buss et al. 1993; Kwok, Hrivnak, & Geballe 1995). Initially, the material ejected during the AGB stage is excited by the remaining, relatively cool, central giant star ($T \sim 5300 \text{ K}$ for this object; Kwok et al. 1995). Eventually, the outer layers of the star are thrown off, exposing the ejecta to the harsh ionizing radiation of the remaining, extremely hot stellar core ($T \sim 50,000\text{--}150,000 \text{ K}$) and initiating the planetary nebular phase. The transition phase lasts on the order of 10^3 yr . The best match to the spectrum of IRAS 22272+5435 (Fig. 3) is produced by a PAH mixture dominated by neutral PAHs ($\sim 60\%$), and it includes species with a broad range of thermodynamic stabilities, from dicoronylene, the most stable PAH in our database, to naphthalene, the least stable. Such a mixture is certainly reasonable considering the nature of this object. In the earliest stage of the nebula, it is logical to expect that the emitting material would contain a diverse mixture of freshly made species, representing a wide range of thermodynamic stabilities, structures, and sizes. Furthermore, given the relatively benign radiation field produced by a 5300 K star, a mixture of neutral and ionized species is likely. The match shown in Figure 3 reflects exactly these characteristics.

Next, let us consider the very different environment represented by the ionization ridge in the Orion Nebula. The Orion spectrum shown in the bottom frame of Figure 3 samples the interface between a cold, dense, molecular cloud and an H II region (Bregman et al. 1989). Here the material produced during the protoplanetary phase has been “aged” for perhaps a billion years in the ISM, having been processed through many diffuse, dense medium cycles where it was alternately exposed

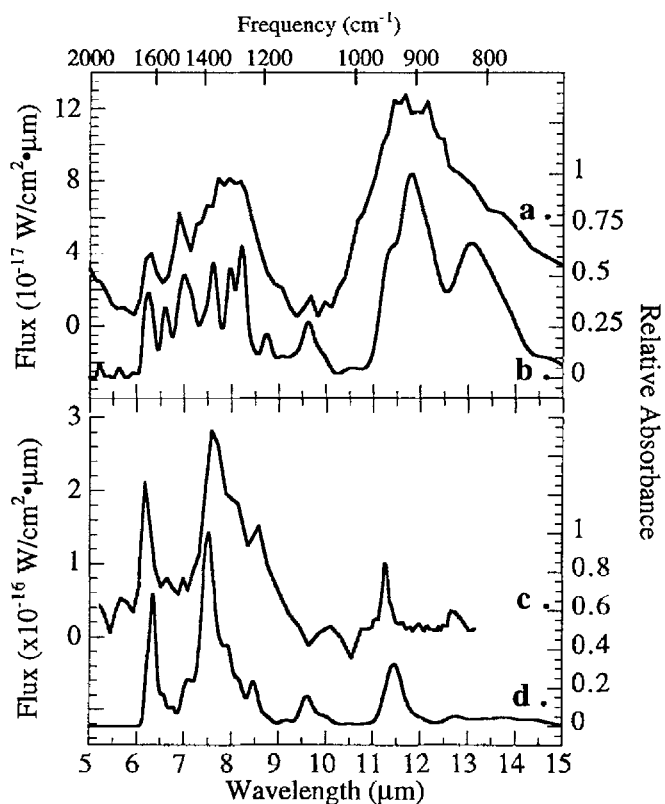


FIG. 3.—*Top*: the (a) *emission* spectrum from the proto-planetary nebula IRAS 22272+5435 compared with the (b) *absorption* spectrum produced from a mixture of neutral and cationic PAHs comprised of 18% dicoronylene⁰; 14% naphthalene⁺ and 9, 10-dihydrobenzo(e)pyrene⁺; 11% 9, 10-dihydrobenzo(e)pyrene⁰; 10% benzo(j)fluoranthene⁰ and coronene⁰; 3% benzo(a)fluoranthene⁺; 3% benzo(j)fluoranthene⁺; 3% coronene⁺; 3% hexabenzocoronene⁺; 3% dicoronylene⁺, benzo(a)fluoranthene⁰, naphthalene⁰, and hexabenzocoronene⁰ (see Fig. 1). The spectrum of IRAS 22272+5435 is reproduced from Buss et al. (1993). *Bottom*: the (c) *emission* spectrum from the Orion ionization ridge compared with the (d) *absorption* spectrum produced from a mixture of fully ionized PAHs consisting of 20% benzo(k)fluoranthene⁺, and dicoronylene⁺; 10% coronene⁺, benzo(b)fluoranthene⁺, 9, 10-dihydrobenzo(e)pyrene⁺ and phenanthrene⁺; 5% benzo(ghi)perylene⁺, tetracene⁺, and benz(a)anthracene⁺; 2% chrysene⁺ and fluoranthene⁺ (see Fig. 1). The Orion spectrum is reproduced from Bregman et al. (1989).

to the very harsh diffuse interstellar radiation field and then “sheltered” in a dense molecular cloud. Now this material is exposed to the ionizing radiation from the adjacent hot young O stars making up the Trapezium. The best match to the Orion spectrum with the spectra in our database is quite revealing about the nature of the PAH population there. Unlike the proto-planetary nebula case, in which a 60%–40% neutral-cation ionization balance is indicated, a completely ionized PAH mixture is indicated for Orion. Furthermore, the less stable PAHs that played a role in the proto-planetary nebula appear to be much less abundant in Orion. Instead, the most thermodynamically stable members of the PAH family dominate the emission. In fact, four of the most stable PAHs in our mixture contribute 60% of the match to Orion. Although the spectra in our database reproduce all the major peaks and relative intensities of the Orion spectrum reasonably well, the 1300 cm^{-1} feature of the co-added laboratory spectrum is significantly narrower than the interstellar band. This reflects the lack of the prominent 1280 cm^{-1} component (Bregman 1989) and the broad emission underlying the 1613, 1310, and 1160

cm^{-1} bands observed in the astronomical emission (Cohen et al. 1986, 1989). This deficiency is likely attributable to emission from a class or classes of species not yet represented in our database and emission from small, solid grains. The nature of the PAH population revealed by the Orion comparison is again precisely what one would expect given the history of this object—an object in which only PAHs that have survived the entire interstellar gauntlet are present. The less stable components of the carbon-rich material that was initially ejected into the ISM during the red giant and proto-planetary nebula phase have long since been “weeded out” by interstellar radiation, cosmic-ray bombardment, incorporation into the giant molecular cloud, and finally erosion by the UV-rich radiation from the nearby, newly formed stars. In addition, since PAH ionization potentials lie between 6 and 8 eV, the survivors should be largely ionized in that radiation field. Thus, it is entirely reasonable that the best-fit PAH mixture for the Orion ionization ridge should reflect a disproportionately large contribution from the hardest species and should be dominated by ionized species.

Free individual PAH molecules, PAH clusters, and particles comprised of aromatic (PAH) subunits are responsible for the overall interstellar emission spectrum. While earlier work suggests that particles produce the underlying continuum at the lower frequencies (Cohen et al. 1986; Allamandola, Tielens, & Barker 1987; Blanco, Bussoletti, & Colangeli 1988; Flickinger & Wdowiak 1990), Figure 3 shows that the individual emission features can be accounted for by free PAH molecules. Within the framework of the PAH model, such molecular-sized PAHs constitute the small end of the distribution of aromatic, rich interstellar materials, with PAHs containing less than 50 carbon atoms (such as those used here) contributing over 95% of the emission at 3050 cm^{-1} , roughly 30% of the 1610 cm^{-1} band, and 20% of the 1310 cm^{-1} complex (Schutte, Tielens, & Allamandola 1993; Allain, Leach, & Sedlmayr 1996). It is striking that such good matches are obtained from a limited

database representing only the very smallest interstellar PAH molecules possible.

Since IRAS 22272+5435 and the Orion ionization ridge represent very different epochs in the evolution of cosmic carbon, the spectral differences reveal how carbonaceous material evolves as it passes from its circumstellar birth site into the general ISM. While there can be great variability in the appearance of the interstellar spectra between objects or from one region to another within one object, these differences can readily and naturally be accommodated by different PAH populations. In turn, the different molecular compositions used for the comparisons reveal differences in ionization balance, chemical composition, stabilities, structures, ionization potentials, electron affinities, and the electron sticking coefficients of the PAHs in the different emission regions. While the PAH mixtures used to provide the spectral fits in the above examples are not unique, only minimal variation is possible in the choice of the dominant PAHs in each. This ability to produce close matches to the interstellar spectra supports the PAH hypothesis and provides an important new probe of a wide variety of interstellar conditions and histories—a probe that can be used across the whole of the ISM, from the regions where stars are born to those where they are dying, and in the intervening diffuse ISM. PAHs hold the promise of being a very important interstellar probe, much like CO has been in the last quarter of the twentieth century.

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