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PERIPHERALLY HYDROGENATED NEUTRAL POLYCYCLIC AROMATIC HYDROCARBONS AS CARRIERS OF THE 3 MICRON INTERSTELLAR INFRARED EMISSION COMPLEX: RESULTS FROM SINGLE-PHOTON INFRARED EMISSION SPECTROSCOPY

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ABSTRACT

Infrared emission spectra of five gas-phase UV laser-excited polycyclic aromatic hydrocarbons (PAHs) containing aliphatic hydrogens are compared with the main 3.3 μm and associated interstellar unidentified infrared emission bands (UIRs). We show that neutral PAHs can account for the majority of the 3 μm emission complex while making little contribution to the other UIR bands; peripherally hydrogenated PAHs produce a better match to astrophysical data than do those containing methyl side groups; 3.4 μm plateau emission is shown to be a general spectral feature of vibrationally excited PAHs containing aliphatic hydrogens, especially those containing methyl groups; and finally, hot-band and overtone emissions arising from aromatic C—H vibrations are not observed in laboratory emission spectra, and therefore, in contrast to current assignments, are not expected to be observed in the UIRs.

Subject Headings: infrared: ISM: lines and bands — ISM: lines and bands — ISM: molecules — molecular data — molecular processes

1. INTRODUCTION

Like the diffuse interstellar bands (DIBs), the unidentified infrared bands (UIRs) are a spectroscopic enigma that have occupied astronomers and spectroscopists for decades. Mounting evidence suggests that the intense infrared emissions at 3.3, 6.2, 7.7, 8.6, and 11.2 μm originate from carbon-containing molecules. The similarities of UIRs to polycyclic aromatic hydrocarbon (PAH) spectra, and the thermodynamic stability of these molecules, underlie compelling arguments in favor of assigning the UIRs to highly excited gas-phase PAHs (Léger & Puget 1984; Allamandola, Tielens, & Barker 1985; Allamandola, Tielens, & Barker 1989). Neutral PAHs present a severe mismatch with UIR emissions with respect to the relative intensities of the bands, while PAH ions have gained credibility based on their low-ionization energies, ab initio results, and cryogenic matrix spectra demonstrating more favorable band intensities and positions (Allamandola et al. 1999).

The most-studied UIR feature near 3.3 μm corresponds to an aromatic C—H stretch. This spectral region is ground-accessible to astronomers (Geballe et al. 1985; deMuizon et al. 1986; Joblin et al. 1996), while infrared detectors are available for gas-phase laboratory studies (Cherchneff & Barker 1989; Flickinger, Wdowiak, & Gómez 1991; Brenner & Barker 1992). However, because all aromatic molecules exhibit this feature, it is not a definitive diagnostic for assignment of the UIRs to a particular carrier. The extension of gas-phase emission studies deeper into the IR made possible by our development of the SPIRES technique (Schlemmer et al. 1994; Cook et al. 1996), has shown that neutral PAHs as large as coronene fail to reproduce the IR emissions in both band positions and relative intensities (Cook et al. 1998).

In many astrophysical sources there are, associated with the 3.3 μm band, several satellite emissions that appear to the red of the main band and with an underlying plateau. Variations in the 3.3 μm band and associated minor features have been measured at varying distances from excitation sources in the Red Rectangle nebulae surrounding HD 44179 and in the Orion bar (Geballe et al. 1989), where emission features were reported at 3.40, 3.46, 3.51, and 3.57 μm, with noticeable shoulders at 3.415 and 3.53 μm and with plateau emission extending from 3.2–3.6 μm. The 3.40 and 3.51 μm emission features were found to be spatially correlated, falling off less rapidly than the other features when moving away from the excitation source, such that their intensities increased relative to the 3.29 μm main feature. The 3.46 and 3.53 μm features were attributed to overtones and combination bands of PAH C=C stretching vibrations, while the 3.40 and 3.51 μm features were ascribed to aromatic C—H stretch hot bands, or to C—H stretching motions of aliphatic side groups. A similar study corroborated these observations, reporting features at 3.292, 3.403, 3.463, and 3.515 μm in NGC 1333 (Joblin et al. 1996). Those authors conclude that methyl side groups on PAHs are primarily responsible for the minor features, while hot bands make only a minor contribution.

In an attempt to clarify whether the 3.40 μm emission arises from aliphatic PAHs or is due to aromatic C—H stretch hot bands, overtone emissions were sought near 1.67 μm as evidence for the former. In one observation (Geballe et al. 1994), an extremely weak emission centered at 1.680 μm was assigned to the aromatic C—H stretch overtone ($\chi = 2 \leftarrow 0$), placing the hot-band emission at 3.43 μm, ostensibly proving that the band centered at 3.40 μm band is not the aromatic C—H stretch hot band, leaving the long-wavelength shoulder of the 3.40 μm band assigned to the $\chi = 2 \leftarrow 1$ transition. They concluded that the 3.40 μm band must therefore arise from aliphatic side groups on PAH molecules. Based on laboratory evidence presented here, we argue that assignment of the 1.680 μm emission to the C—H overtone is probably in error, and none of the

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satellite features observed in the 3.3 μm emission spectrum are due to hot-band transitions.

Bernstein et al. compared a number of H_n-PAHs (peripherally hydrogenated PAHs) and one methylated PAH (methyl-coronene) to emission from the Orion bar and IRAS 05431−0852, a more benign environment (Bernstein, Sandford, & Allamandola 1996). It was shown that complete excess hydrogen coverage resulted in an absorption spectrum that better matched the *IRAS* source, while low excess hydrogen coverage was consistent with regions of greater photon flux in the Orion bar. Apparently, no plateau feature was observed in those cryogenic matrix absorption studies, so the origin of that feature remained unclear.

In this paper we present new SPIRES spectra recorded under conditions that closely simulate the astrophysical environment thought to produce the UIRs (viz., highly excited, isolated, neutral PAHs, recorded in emission). We investigate both methyl-substituted and peripherally hydrogenated PAHs near 3.3 μm and compare the results with high-resolution IR emission spectra from IRAS 21282+5050.

2. RESULTS AND DISCUSSION

The Berkeley Single Photon InfraRed Emission Spectrometer (SPIRES) is diagramed in Figure 1 and has been described in detail before (Cook et al. 1998). The five samples examined in this study, 2-methylnaphthalene (98%), 2-methylphenanthrene (98%), 1,2,3,6,7,8-hexahydropyrene (98%), 9,10-dihydrophenanthrene (94%), and acenaphthene (97%), were purchased from Aldrich and were used without further purification. Spectral features are fitted to multiple Gaussians using a cubic polynomial peak fitting routine. Peak positions, widths, and amplitudes are obtained, but may not be directly comparable to published astronomical observations, where plateau emission is treated as the baseline value in some reports. Table 1 presents the details of the 3.3 μm, satellite, and plateau emission features observed for the five PAH molecules studied in this work.

An example of the fitting procedure is illustrated for 2-methylnaphthalene (7 aromatic H, 3 aliphatic H) in Figure 1. This molecule shows several distinct peaks in its emission spectrum whose convolution do not account for the under-

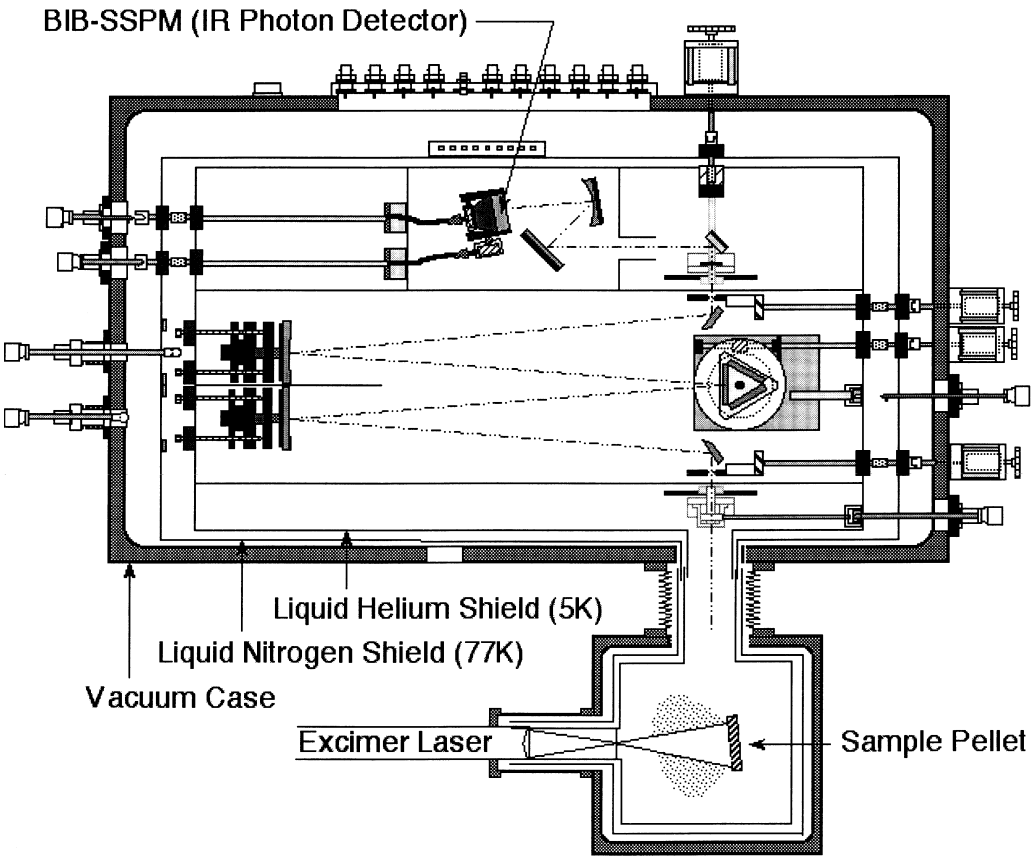

FIG. 1. Berkeley Single Photon InfraRed Emission Spectrometer (SPIRES). Highly excited PAHs are generated inside a dark cryogenic vacuum chamber via laser desorption from a pellet of pure PAH affixed to a 77 K copper target. The minimum laser pulse energy required to generate detectable signal without burning the sample or fragmenting the molecules (typically 30–100 mJ pulse^{-1} in^2 unfocussed 248 nm excimer laser radiation) is employed. The resulting gaseous molecules traverse the viewing region of a liquid helium cooled monochromator to which the sample chamber is connected via light-tight baffles. Each laser pulse generates a plume of molecules whose IR emission signal lasts for about 100 μs. The light emitted is dispersed in the cryogenic (5 K) 0.5m f/7 Czerney-Turner monochromator (Infrared Laboratories), which is outfitted with three diffraction gratings, and filter wheels at the entrance and exit slits for order selection and background/stray light suppression, allowing for complete spectral coverage of the UIR spectral region (2–16 μm). Light from the exit slit is imaged onto a vertical 10 element array Blocked Impurity Band Solid State Photomultiplier (BIB-SSPM, Rockwell International; Petroff, Stapelbroek, & Kleinhans 1987) held at 7 K. This device is capable of detecting single photons from 420 nm–28 μm, but suffers from an extremely limited dynamic range, necessitating the cryogenic environment. A bank of amplifiers and discriminators convert IR photon counts into digital pulses, and transient signals are recorded on a multichannel scalar (Stanford Research SR430), where 5–10 successive desorption events are summed and transferred to a Macintosh G3 computer for display and analysis.

TABLE 1

SPIRES Features for the Five H-PAHs Studied in this Work

Apparent Peak Positions		Fitted Peak Positions			
		Positions			
Position (μm)	FWHM (cm^{-1})	(μm)	cm^{-1}	FWHM (cm^{-1})	Relative Intensities
2-Methylnaphthalene					
3.28	79	3.276	3051.8	44.2	1.00
		(3.319)	(3012.2)	(22.2)	0.236
		(3.344)	(2990.1)	(18.7)	0.088
		(3.370)[a]	(2967.2)	(180.4)	0.327
3.42		3.423	2921.1	30.0	0.246
3.47		3.476	(2876.6)	(27.3)	0.124
		(3.501)	(2856.3)	(16.6)	0.059
2-Methylphenanthrene					
3.29	77	3.270	3058.1	53.6	1.00
		3.316	3015.4	57.3	0.789
3.42		3.426	2918.3	26.8	0.212
		(3.418)[a]	(2925.3)	(202.0)	0.672
3.51		3.514	2845.9	6.8	0.072
1,2,3,6,7,8-Hexahydropyrene					
		(3.249)	(3078.3)	(29.7)	0.075
3.29	32	3.296	3034.2	39.0	0.329
3.40	20	3.395	2945.6	29.3	1.00
		(3.466)[a]	(2885.5)	(87.9)	0.239
3.52		3.521	2840.18	12.0	0.120
9,10-Dihydrophenanthrene					
3.27	90	3.261	3038.0	53.7	0.829
		3.319	3012.5	62.3	0.697
3.40	26	3.406	2936.3	37.42	1.00
3.46		3.455	2894.7	29.1	0.610
3.53		3.514	2845.7	59.0	0.495
Acenaphthene					
3.25	53	3.246	3080.3	52.0	0.867
3.38	44	3.384	2955.5	34.8	1.00
		(3.415)[a]	(2928.0)	(148.9)	0.198
3.50		3.499	2957.4	27.43	0.110

Note.—Apparent peak positions are determined by visual inspection and tabulated to facilitate comparisons to astrophysical data. Calculated peak positions are determined from a cubic polynomial multipeak fit to determine peak positions, widths, and relative intensities. The relative intensities are determined from the fitted values relative to the baseline and not the apparent intensities relative to the plateau emission. This may differ from the way some astrophysical data are presented in the literature. Relative intensities are separately normalized to the dominant peak in each spectrum, and are not comparable between different molecules. Values in parentheses are shoulders on the main peaks, plateau emissions, or otherwise discernible but less distinct features required to obtain a reasonable fit to the data.

[a] Fitted plateau emission.

lying plateau emission. By including a broad band to better reproduce the plateau, excellent fit residuals are obtained. The spectrum is dominated by a peak at 3.28 μm that is a blended band with clearly discernible features at 3.276 (aromatic C—H stretch), 3.319, and 3.344 μm. The minor features at 3.42 and 3.47 μm can be assigned to the aliphatic C—H stretch of the methyl group, while the other minor features, shoulders, and plateau are most likely combination bands. For comparison, the SPIRES spectrum of pyrene is included to show the extremely weak (often absent) plateau emission exhibited by "normal" PAHs. All of the emission spectra in this work are plotted together against astrophysical data in Figure 2.

2-methylphenanthrene (9 aromatic H, 3 aliphatic H) exhibits a similar plateau emission with several discernible features. The complexity of the emission precluded unambiguous fitting. However, including a broad plateau improved the fit. As for 2-methylnaphthlene, the second most prominent feature (3.426 μm, the aliphatic C—H stretch) occurs red of the UIR emission at 3.40 μm. This sample also exhibits a broad ($\sim$200 cm^{-1}) plateau emission extending from 3.2–3.6μm, resembling the plateau of UIR emitting regions. In methylphenanthrene, this emission is most likely due to the convolution of aromatic and aliphatic C—H stretching modes and combination bands that are broadened as a result of excitations in lower frequency

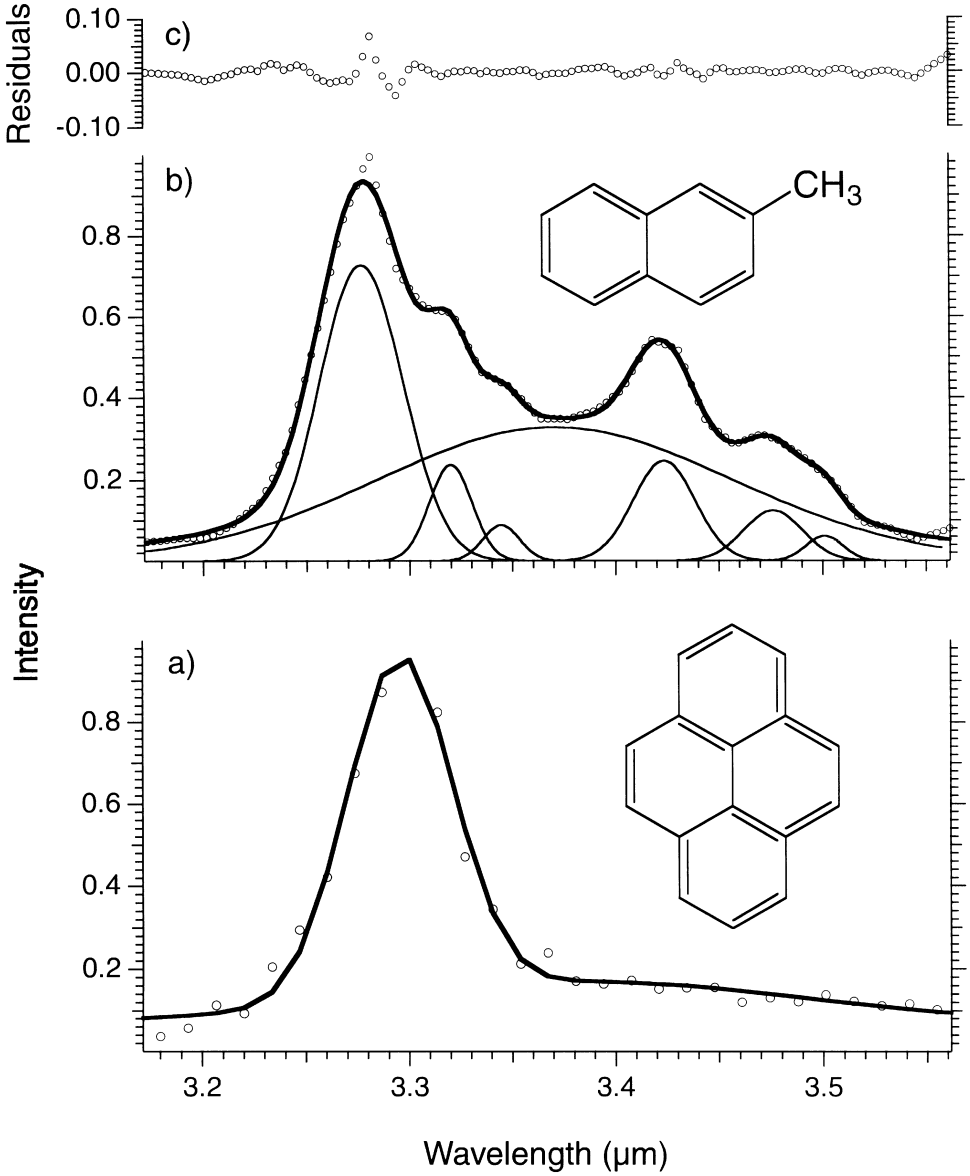

FIG. 2. (*a*) Pyrene SPIRES spectrum demonstrating the absence of a distinct plateau emission from a "normal" (nonhydrogenated) PAH. Small PAHs do exhibit a weak red tail on the 3.3 μm emission, barely evident in this spectrum, which rapidly diminishes for larger PAHs. (*b*) 2-methylnaphthalene emission spectrum (*open circles*) fitted to seven Gaussians. The solid line drawn through the data is the sum of the fitted peaks at the data *x*-values. Note the plateau emission not accounted for by the sum of the aliphatic C—H features red of 3.3 μm, and (*c*) the fit residuals.

modes (combination or sequence bands; Cook et al. 1998). However, other processes cannot be ruled out. For instance, a high degree of internal rotation of the methyl group could lead to a vibrational band with a severely rotationally broadened profile. Another possibility is that the aliphatic C—H bend is strongly coupled to excited low-frequency ring modes, enhancing and broadening its emission. Such plateau emission seems to be a general feature of highly excited PAHs with aliphatic hydrogens and is an especially prominent feature of the methylated PAHs studied here.

It was previously proposed that methyl groups are mainly responsible for the 3.4 μm band, based primarily on studies of methyl-coronene (Joblin et al. 1995). However, our results corroborate recent matrix isolation data (Bernstein et al. 1996) that implicate peripherally hydrogenated PAHs as more likely matches to astrophysical observations. It is clear that the minor bands observed for 2-methylnaphthalene and 2-methyphenanthrene, like those of methylcoronene, fall slightly to the red of the UIR emission feature at 3.40 μm and are therefore not the best candidates for the carriers of this band. However, this feature does have a shoulder at 3.41 μm to which methylated PAHs could contribute.

Hexahydropyrene (4 aromatic H, 12 aliphatic H) has a 3 μm emission dominated by the aliphatic C—H stretch at 3.395 μm, which is 3 times stronger than the aromatic C—H stretch at 3.296 μm. One other prominent feature at 3.521 μm is clearly discernible, and like the methylated PAHs studied, a broad plateau is observed but is found to be much narrower ($\sim$90 cm^{-1}), such that the dip at 3.35 μm goes nearly to the baseline. Again, the origin of the plateau emission is unclear, although for this molecule it exhibits significant structure, suggesting that the plateau is a convolution of combination bands originating from C=C stretching modes in the 1600–1400 cm^{-1} region. Additionally, a shoulder is observed at 3.249 μm on the blue edge of the aromatic

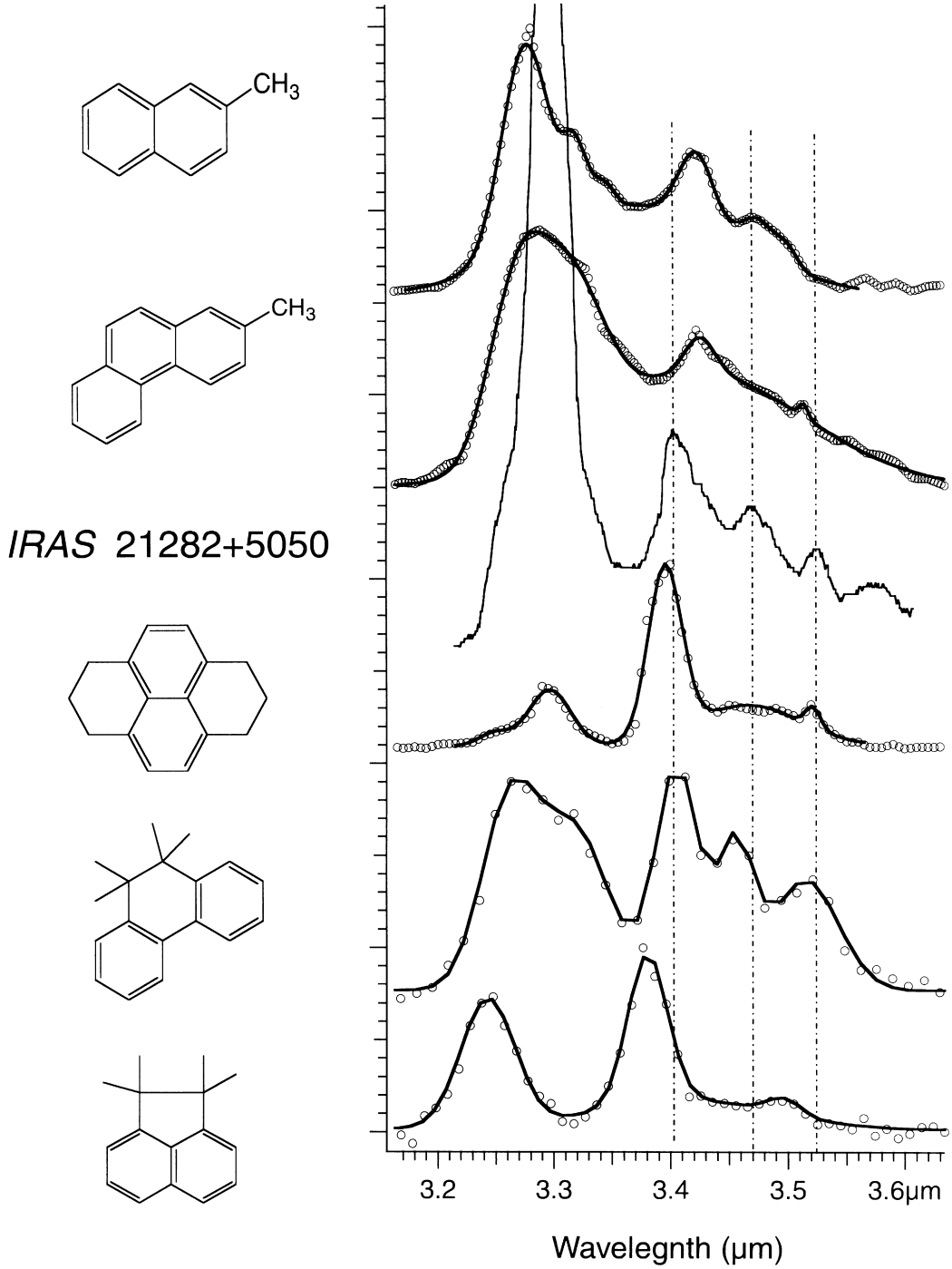

FIG. 3. Five neutral PAHs containing aliphatic hydrogens are plotted against emission features emanating from *IRAS* source 21282+5050 (reproduced from deMuizon et al. 1986). The intensity scale is arbitrary. Open circles represent laboratory data; solid lines through the laboratory data are the sum of Gaussian peak fits as described in the text and illustrated in Fig. 1. Vertical dotted lines are drawn through the minor features in the astrophysical data as a guide to the eye. The peripherally hydrogenated PAHs are clearly a better match to the astrophysical spectrum than are methylated PAHs. Note, however, that the dominant feature in the interstellar spectrum dwarfs the minor bands, which dominate the laboratory spectra of the peripherally hydrogenated PAHs in this study.

C—H stretch feature, similar to the shoulder observed in nearly half of UIR emitting sources (see, for example, de Muizon, Cox & Lequeux 1990). This feature is a common but normally weak feature of many aromatic structures (Bellamy 1975). The peak at 3.395 is a close match to the most prominent UIR satellite feature (3.40 μm), while the 3.521 μm band falls near the 3.51 μm UIR feature found to spatially correlate with the 3.40 μm feature (Geballe et al. 1989). The relative intensities of the minor features is far too great compared to the 3.3 μm band to compare favorably to the UIRs, due to the intrinsically greater oscillator strength of the aliphatic C—H stretch and the high ratio (12:4) of aliphatic to aromatic hydrogens present. However, as the number of aromatic moieties will determine the relative strength of the 3.3 μm band (Bellamy 1975), one could certainly imagine a much larger PAH structure containing

only one such hydrogenated ring whose relative band intensities better match the UIR features (Bernstein et al. 1996; Sloan et al. 1997).

There are five prominent emissions from 9,10-dihydrophenethrene (8 aromatic H, 4 aliphatic H) in the 3.3 μm spectral region: a blended feature comprised of two bands at 3.261 and 3.319 μm, and three peaks of decreasing intensity at 3.406, 3.455, and 3.514. The absolute positions of these three features present a remarkably good match to the emission spectrum of the Orion bar (Geballe et al. 1989), and IRAS 21282+5050 (deMuizon et al. 1986). Though a relative intensity mismatch exists for this sample, it should be noted that the samples studied here are much smaller than the PAH molecules proposed to give rise to the UIR features ($\geq$30–50 carbons) at this wavelegnth, while the structure of dihydrophenathrene can be viewed as a fragment or edge structure of a larger and more condensed structure like dihydrocircumcoronene ($C_{54}H_{20}$; Fig. 3). There is also an apparent plateau emission in the spectrum, but in this case it is adequately represented by the convolution of the fitted peaks (i.e., without explicitly including a broad plateau).

Acenapthalene (6 aromatic H, 4 aliphatic H) has received attention in the literature within several astrophysical contexts, being described as a keystone species in the cycle of PAH formation, implicated in the UV extinction bump, and within the UIR PAH hypothesis (Beegle et al. 1997). It is characterized by having additional hydrogen coverage on a five membered ring in the PAH structure. Its peaks at 3.246, 3.384, and 3.499 occur blueshifted from UIR features, and therefore are not a particularly good match to astrophysical emissions. A weak plateau emission is also apparent in its spectrum.

3. CONCLUSIONS

3.1. *The 3.4 μm Satellite Complex*

The 3.40, and 3.51 μm interstellar emission features are well matched by laboratory emission spectra of hexahydropyrene and 9,10-dihydrophenanthrene, and a 3.46 μm feature is also observed in 9,10-dihydrophenethrene, making that molecule the best explicit match to the observed 3 μm data thus far measured in the laboratory. We conclude that peripheral hydrogen coverage on PAHs with primarily two adjacent carbons on the ring edge (e.g., coronene) are better candidates for the emission than are PAH molecules having three or more adjacent carbons on a ring edge (e.g., naphthalene, pyrene). Hydrogenation on five-membered rings does not yield a favorable match. Methyl groups attached to PAH molecules are a poorer match to the astrophysical data, although they may contribute to shoulders observed on the more prominent minor features.

3.2. *Plateau Emission from Highly Excited Hydrogenated and Methylated PAHs*

The 3.3 μm UIR bands are often accompanied by a broad plateau extending from 3.2–3.6 μm. Allamandola et al. assigned this to a vibrational quasi-continuum from overlapping C—H stretching vibrations, overtones and combination bands of C=C stretching fundamentals from the 5.5–9.5 μm regions, and C—H stretches of aliphatic side groups (Allamandola et al. 1989). However, to our knowledge no laboratory evidence has corroborated these assignments of the plateau until now.

The plateau emission can indeed be understood in terms of the convolution of emissions from both aromatic and aliphatic C—H stretches of PAHs. In fact, we deduce from our experiments that 3.4 μm plateau emission appears to be a general *emission* feature of highly excited methylated and H_n-PAHs while appearing only weakly in the spectra of small "normal" PAHs. A plateau is not apparent in the *absorption* spectra of cold molecules (Bernstein et al. 1996). While a 3.4 μm plateau can be observed quite clearly in the elevated temperature spectrum of 1-methylcoronene (Colangeli, Mennella, & Bussoletti 1992), it is difficult to separate contributions from hot bands in the absorption spectrum.

3.3. *Hot-Band and Overtone Emissions*

It has long been speculated that the 3.40 and 3.51 μm UIR bands are due to anharmonically shifted aromatic C—H stretch hot bands, while the other minor features are overtones of C=C stretching modes (Allamandola et al. 1989). Astronomical observation of an extremely weak feature at 1.68 μm was attributed to the $\chi = 2 \leftarrow 0$ overtone of the aromatic C—H stretch (Geballe et al. 1994), placing the hot band at 3.43 μm, red of the 3.40 μm peak. This has been presented numerous times as definitive proof that the 3.40 μm feature is not a hot band.

In the PAH-UIR model, internal conversion from a UV-pumped electronic excited state to the ground state results in a random distribution of the excitation energy among many vibrational modes. For this top-down excitation, it is statistically improbable for large molecules to have multiple quanta in any one vibrational mode (especially higher energy modes). Furthermore, the 3.40 μm band tends to grow in intensity relative to the 3.29 μm feature moving

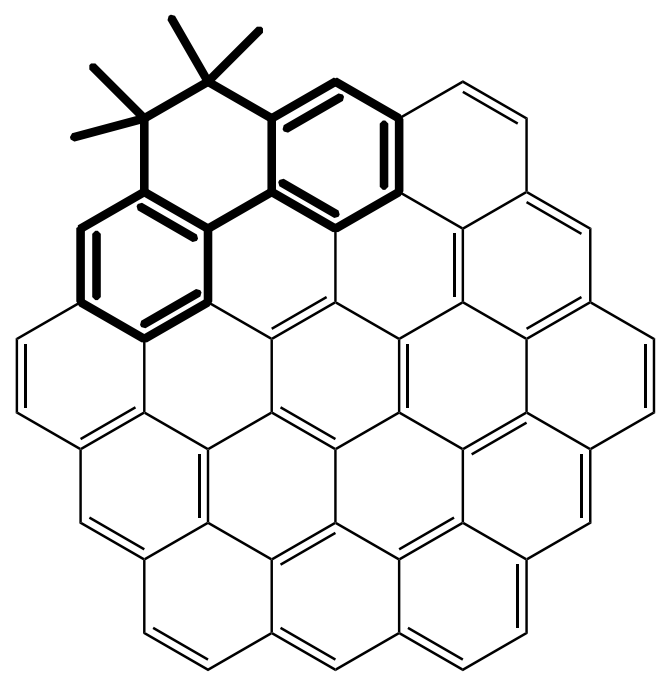

FIG. 4. Dihydrocircumcoronene ($C_{54}H_{20}$) has an edge structure like 9,10-dihydropheneanthrene, which is shown to be an excellent match to the UIR emission with respect to band position, but not the astronomical relative intensities. Neutral peripherally hydrogenated PAHs of this size will better match the astrophysical data and could account for most of the emission in the 3.3 μm region of the spectrum while making little contribution to lower frequency emission features.

away from the exciting star. This disfavors assignment to hot bands, because as the radiation field and average photon energy decrease, so should hot band and overtone emissions. Finally, our data for PAHs as large as coronene show no evidence for hot bands or overtone emission for any vibrational mode between 2–16 μm (Cook et al. 1996). While it cannot be ruled out that combination bands and overtones are responsible for some of the minor features of methylated/hydrogenated PAHs, it seems that these processes are not important for normal PAHs. These observations call into question assignment of the 1.68 μm emission to an aromatic C—H overtone and furthermore do not support the proposal that any 3.3 μm feature arises from aromatic C—H hot bands (Geballe et al. 1994). A spatially correlated astronomical study of possible overtone and hot bands will be required to confirm that assignment.

3.4. *Neutral PAHs as Carriers of the 3.3 μm Emission Complex*

No PAH emission spectrum has been able to reproduce the UIR spectrum with respect to either band positions or relative intensities (Cook et al. 1998). Most notably, the 3.3 μm band is far too intense in neutral PAHs, wherein it dominates the spectrum of most samples studied (Cook et al. 1998). Ionized PAHs are currently thought to better match the UIR band relative intensities, and hence have become the favored candidates for the UIR carriers. However, ab initio results show that IR band strengths for the aromatic C—H stretch become vanishingly small, whereas the 3.3 μm feature ranges from 4% to 25% of the "7.7" μm emission complex in the astrophysical spectra, suggesting that ions cannot be fully responsible for the 3.3 μm emission found in many sources. In fact, essentially all of the 3.3 μm UIR emission from some sources could be accounted for by neutral PAHs, which would then make negligible contribution to the 6–9 μm features (dominated by ions). Spatially resolved astronomical observations comparing the 6–9 μm intensities to the 3 μm bands as a function of local environment will be helpful in elucidating this possibility. Nevertheless, the data presented here suggest that the best candidates for the 3 μm emission carriers in terms of charge state, size, and degree of hydrogenation are molecules like that shown in Figure 4.

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