THE ULTRAVIOLET AND VISIBLE SPECTRUM OF THE POLYCYCLIC AROMATIC HYDROCARBON $C_{10}H_8{}^+$: POSSIBLE CONTRIBUTIONS TO THE DIFFUSE INTERSTELLAR BANDS AND TO THE ULTRAVIOLET–VISIBLE EXTINCTION

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ABSTRACT

This report describes the properties of the cation of the PAH naphthalene ($C_{10}H_8{}^+$) isolated in inert gas matrices under conditions relevant to astrophysical environments. As with other PAH cations, $C_{10}H_8{}^+$ absorbs in the visible. $C_{10}H_8{}^+$ discrete absorption bands fall at 6741, 6520, 6376, 6314, 6151, 5965, 5848, 5784, 5658, 5503, 5341, 5114, and 4560 Å. The band at 6741 Å is the strongest and falls close to the weak 6742 Å DIB. Five other weaker bands (6376, 6314, 5848, 5784, and 5503) also fall remarkably close to the positions of known diffuse interstellar bands (DIBs). The possibility that $C_{10}H_8{}^+$ is responsible for some of the diffuse interstellar bands can be tested by searching for new DIBS at 6520, 6151, and 5965 Å, other strong naphthalene cation band positions. If $C_{10}H_8{}^+$ is indeed responsible for the 6742 Å feature, it accounts for 0.3% of the cosmic carbon. We also report a remarkably intense, but puzzling, very broad continuum extending from the UV to the visible (2000–3800 Å, with a weak tail extending to ≈ 5100 Å) which seems to be associated with the ion. The molar absorption coefficient (ϵ) at the peak of the continuum is 2.0×10^6 dm^3 mol^{-1} cm^{-1} ($\sigma_e = 7.6 \times 10^{-15}$ cm^2 molecule^{-1}). If a continuum is a general property of PAH cations, this characteristic will have a strong impact on our understanding of how PAHs convert interstellar UV and visible radiation into IR radiation. We also note that neutral naphthalene may contribute to the 2200 Å bump. To produce a detectable feature at 2116 Å, however, requires a high abundance.

Subject headings: ISM: molecules — line: identification — molecular data

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) and related aromatic materials are thought to produce the well-known family of interstellar emission bands between 3300 and 700 cm^{-1} (3 and 14 μm) (Duley & Williams 1981; Léger & Puget 1984; Sakata et al. 1984; Allamandola, Tielens, & Barker 1985; Blanco, Bussoletti, & Colangeli 1988). To account for the observed intensities requires that the carriers of the bands must be small enough (< 10 Å) (Allamandola, Tielens, & Barker 1989; Puget & Léger 1989) and must absorb over a wide energy range—from ultraviolet (UV), to the visible and, in some cases, the near-infrared (NIR) (Aitken & Roche 1983; Sellgren 1984; Sellgren, Luan, & Werner 1990; Désert, Boulanger, & Puget 1990). PAH molecules are extremely stable and present a plausible scheme for the conversion of ultraviolet energy into discrete infrared emission bands in a collision-free medium (through infrared fluorescence). In several high-excitation objects such as the Orion bar, a size distribution of only neutral PAHs can account for the emission. In other cases, such as reflection nebulae (e.g., NGC 2023) and the IR cirrus, small PAHs are required. However, small neutral PAHs absorb only in the ultraviolet (Clar 1964) and, if present, can account only for a fraction of the UV/visible to IR energy transfer needed. In many objects, a large fraction of the PAHs are expected to be ionized (Allamandola et al. 1985) and thus to absorb also in the visible and NIR (Shida & Iwata 1973). Moreover, the ubiquitous presence of ionized PAHs in the interstellar medium make them attractive candidates for the diffuse interstellar absorption bands (DIBs) which extend from about 4400 Å to the near infrared (Van der Zwet & Allamandola 1985; Léger & d'Hendecourt 1985; Crawford, Tielens, & Allamandola 1985).

Unfortunately, very few spectroscopic data are available on PAH ions, data which are needed to address these important pending astrophysical problems. The few data available come mainly from absorption spectra of neutral and ionized PAHs suspended in strongly perturbing media (solid phase [Shida & Iwata 1973; Andrews, Kelsall, & Blankenship 1982] or solution [Delcourt & Rossi 1982]) or from gas-phase photoelectron spectra which do not provide information on all the possible optical transitions (Clark, Brogli, & Heilbronner 1972).

Considering the need for reliable, quantitative spectroscopic data on neutral and ionized, isolated PAHs to test the existing theories and their implications, we have initiated a systematic program to investigate their UV–visible–NIR spectroscopic properties. The first study of this program considers the case of the smallest PAH, naphthalene ($C_{10}H_8$). The detailed spectroscopic analysis of isolated $C_{10}H_8$ and its positive ion ($C_{10}H_8{}^+$) is reported elsewhere (Salama & Allamandola 1991a, b). We discuss, here, the aspects of this study which are relevant to astrophysical applications. In particular, we discuss the possible contribution of $C_{10}H_8{}^+$ to the DIBs and we describe the broad, structureless continuum, extending from 2000 to 5100 Å, which appears associated with the formation of the ion, $C_{10}H_8{}^+$, and which could play an important role in the conversion of interstellar UV and visual radiation to the IR.

2. EXPERIMENTAL

A computer-controlled UV–visible–NIR spectrometer system, coupled to a helium-cooled cryogenic cell, was designed and developed to measure the absorption and emission spectra of neutral, ionic, or radical species in the 1800–9000

Å range under conditions relevant to astrophysical environments. A thorough description of the experimental apparatus and technique is reported elsewhere (Salama & Allamandola 1991a), and only a synopsis will be given here.

Matrix isolation spectroscopy (MIS) is particularly well adapted to astrophysical applications because it involves the trapping of the species of interest ($C_{10}H_8$, $C_{10}H_8^+$) in a chemically inert, rigid cage (rare gas matrix) at low temperature. A gas mixture of naphthalene diluted in the inert gas (Ar or Ne), is condensed onto a (4–20 K) substrate. The frozen matrix is then spectroscopically analyzed. Ions are generated in situ from the stable precursor, $C_{10}H_8$, via vacuum-ultraviolet photoionization.

Briefly, the experimental apparatus, designed for matrix isolation spectroscopy (MIS), consists of a cryogenic sample chamber which is part of an ultrahigh vacuum system and consists of four ports at 90° and two gas deposition ports at 45°. The cryogenic sample holder suspended at the center of the chamber is cooled down to 4.2 K by a variable temperature, liquid helium–flow cryostat. The substrate (sapphire window) can be rotated through 360° without breaking the vacuum and can be positioned to face alternatively the spectroscopy ports, the gas injection ports, an excitation lamp, or vacuum deposition furnace. The spectral light sources consist of a 1600–3600 Å output deuterium lamp for UV spectroscopy and a 3200–25,000 Å output tungsten filament lamp for visible–NIR spectroscopy. The microcomputer-driven monochromator is equipped with two interchangeable gratings blazed at 2500 Å and 5000 Å for UV and visible–NIR spectroscopy, respectively. The ionization source is a microwave-powered, flow discharge hydrogen lamp generating photons of 10.2 eV energy (Lyman-α line). The photon detector consists of a photomultiplier tube (PMT) sensitive in the spectral range 1600–9000 Å and mounted directly on one of the spectroscopy ports of the sample chamber. The PMT analog output signal is transmitted to the microcomputer interfaced to the spectrometer through a NuBus board for digitization and analysis.

An important requirement for astrophysical applications is to use the *least perturbing medium* as matrix material. This is necessary in order to minimize the perturbation of the absorption and emission spectra of the isolated species which would induce a shift in peak positions with respect to the gas phase values making it impossible to compare directly laboratory measurements with astrophysical spectra. Thus, Ne is preferred since it is the least polarizable rare gas material with the highest ionization energy and hence can be used to trap ions with minimum perturbation to the electronic spectrum (Bondybey & Miller 1983; Maier 1989).

3. RESULTS AND DISCUSSION

3.1. *Neutral Naphthalene*

Figure 1 shows the 1800–3600 Å spectrum of *neutral* naphthalene isolated in a Ne matrix. In the wavelength range covered in our experiments (1800–9000 Å), naphthalene shows three distinct band systems with very different intensities (Salama & Allamandola 1991a). System I, peaking at 3076 Å, is very weak [gas-phase oscillator strength (f) = 0.002 (George & Morris 1968)]. System II is moderately strong ($f = 0.1$ [George & Morris 1968]) and peaks at 2685 Å. System III is very strong ($f = 1.0$ [George & Morris 1968]) and peaks at 2116 Å. From the astrophysical perspective, this spectrum indicates the following:

1. Neutral naphthalene does not absorb in the visual and therefore cannot contribute to the DIBs.
2. Despite the fact that the spectrum of naphthalene does not peak at the interstellar 2175 Å absorption, it could contribute, if present, as substructure to the hump of the extinction curve. The cross section of the 2116 Å band (4×10^{-16} cm^2
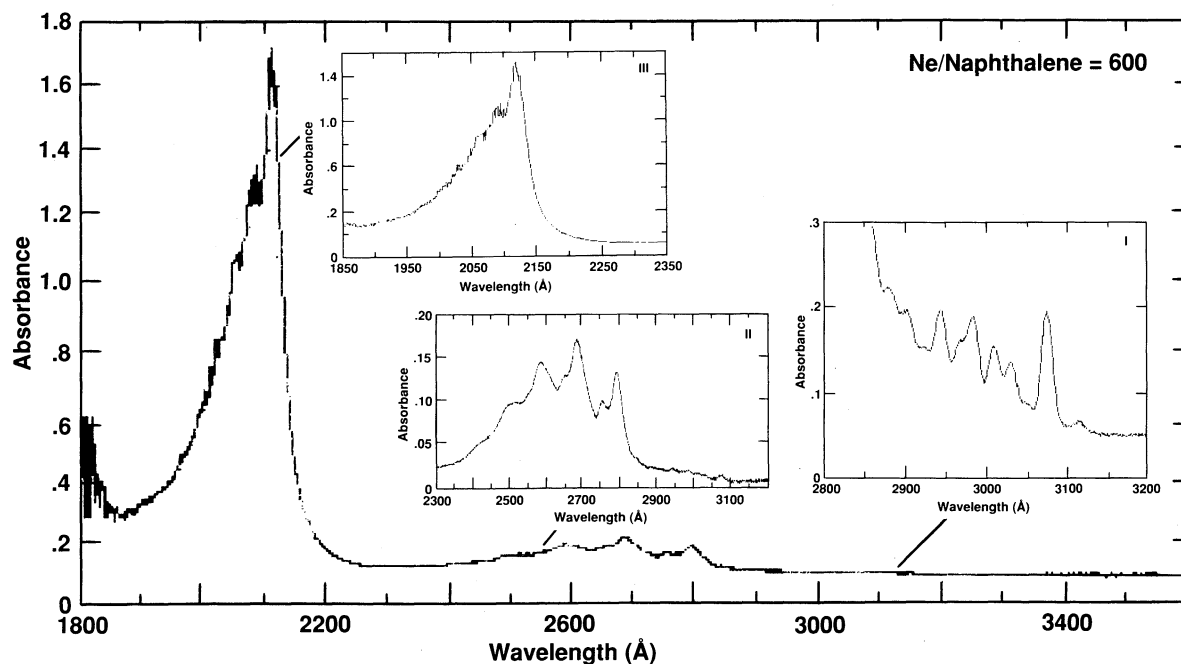

FIG. 1.—The absorption spectrum of neon-matrix isolated naphthalene (Ne/$C_{10}H_8$ = 600, T = 4.2 K) taken from Salama & Allamandola (1991a). The full spectrum corresponds to a 10 minute deposition at a rate of 10 mmole h^{-1}. Inserts I, II, and III correspond to 120, 20, and 10 minute depositions, respectively.

molecule^{-1} [George & Morris 1968]), would require a naphthalene abundance of 1.3×10^{-7} with respect to hydrogen in order to produce detectable substructure on the 2200 Å hump at the 10% level. This abundance is very high, comparable to the overall PAH abundance which is estimated in the range 2.0×10^{-7} (Allamandola et al. 1985) to 5.4×10^{-7} (Puget & Léger 1989).

3.2 The Naphthalene Cation

VUV irradiation of neutral $C_{10}H_8$ isolated in a Ne matrix with 10.2 eV photons produces the naphthalene *cation* ($C_{10}H_8^+$) by direct, one-photon ionization. The new spectral features associated with the cation that arise upon VUV irradiation (Figs 2 and 3) fall into seven band systems (not all shown here) peaking at 6741, 4560, 3769, 3050, 2760, 2443, and 2227 Å respectively and an apparent continuum (Salama & Allamandola 1991a, b). The visible–near-IR system at 6741 Å consists of several discrete bands. The astrophysical implications of the discrete features will first be considered, followed by a discussion of the continuum.

1. Contrary to its neutral precursor, *ionized naphthalene* ($C_{10}H_8^+$) *absorbs at discrete frequencies in the visual and thus could contribute to the DIBs.* In a previous study, the strongest DIB features near 6700 Å appeared to resemble the relative
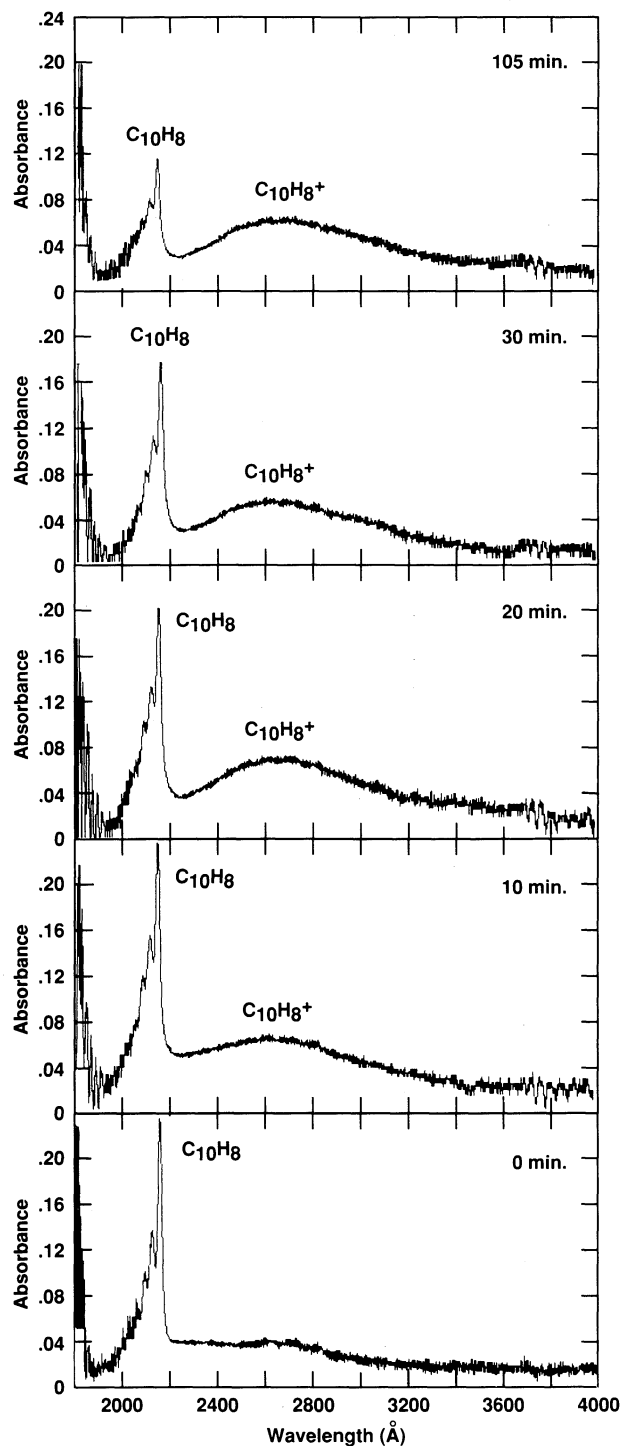

Fig. 2.—The growth of $C_{10}H_8^+$ and simultaneous depletion of the neutral precursor ($C_{10}H_8$) absorptions as a function of photolysis time in an argon matrix at 4.2 K. Starting concentration: $Ar/C_{10}H_8 = 600$.
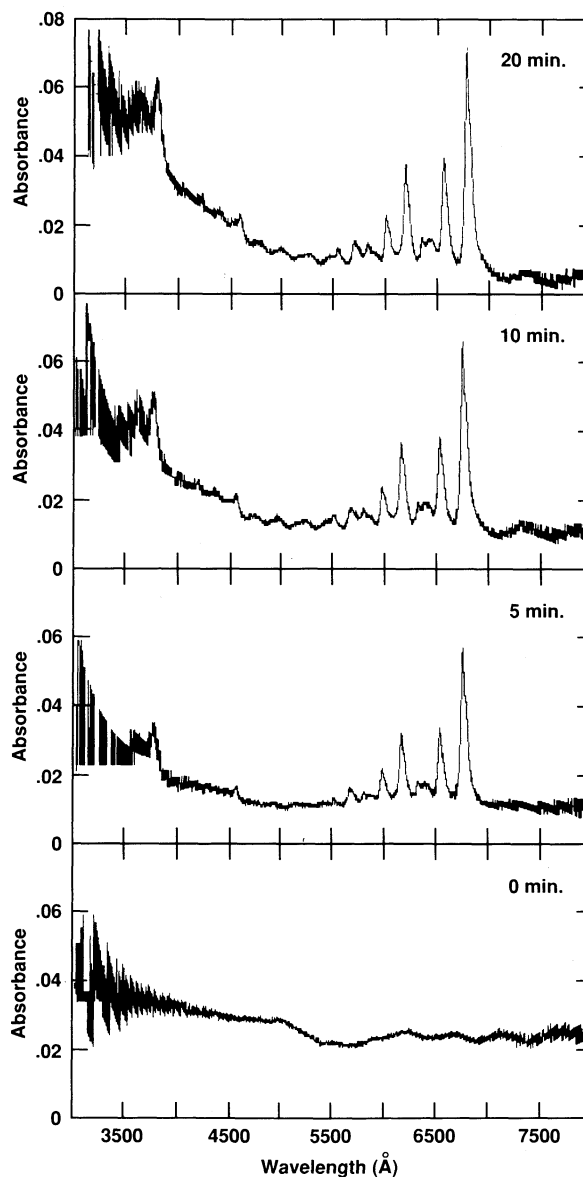
Fig. 3.—The growth of $C_{10}H_8^+$ absorption as a function of photolysis time in a neon matrix at 4.2 K. Starting concentration: $Ne/C_{10}H_8 = 800$.

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intensity pattern of the three most intense bands of the naphthalene cation (Crawford et al. 1985). Unfortunately, this comparison was, then, limited by the fact that the available PAH ion spectra were all measured in severely perturbing media (i.e., organic salts) known to induce a rather strong shift of the intrinsic electronic bands of the trapped species. We now revisit this issue in view of the new laboratory data in a neon matrix which does not perturb the bands significantly.

Table 1 compares the spectral properties of the DIBs (Herbig 1975, 1988) with those of the "unperturbed" naphthalene cation (i.e., isolated in a Ne matrix) in the 5000–7000 Å region (Salama & Allamandola 1991a). *The $C_{10}H_8^+$ bands*

show, in some cases, a remarkable agreement with some of the weaker DIBs.

There are two ways in which one can examine the possible contribution of the naphthalene cation to the DIBs. One extreme is to search for *all* of the $C_{10}H_8^+$ bands, regardless of their intensity, among the DIBs. Another approach is to only require that the strongest $C_{10}H_8^+$ band(s) be present. The former *requires* a high fractional naphthalene abundance among the PAHs; the latter, a relatively lower abundance.

The first approach shows that, while several $C_{10}H_8^+$ bands have peak positions which fall within 5 Å (the resolution element of our spectrometer) of DIBs, there is a relative inten-

TABLE 1

COMPARISON OF $C_{10}H_8^+$ ABSORPTION BANDS WITH THE DIBs IN THE 7000–5000 Å RANGE

DIBs[a]	Equivalent Width (Å)[a]	$C_{10}H_8^+$ (in Ne)[b]	DIBs[a]	Equivalent Width (Å)[a]	$C_{10}H_8^+$ (in Ne)[b]
4882.............	1.27; 0.24; ...; 2.30	...	6113.............	0.07	...
... .	...	5114 (w)	... .	...	6151 (s)
... .	...	5341 (w)	6177.............	1.85; ...; 0.09; 1.70	...
5362[c]............	0.15; ...; ...; 0.20	...	6196[c]............	0.10; ...; 0.03; 0.14	...
5404.............	0.07	...	6203[c]............	0.43; 0.08; 0.06; 0.38	...
5420.............	0.18	...			
5449[c]............	0.56; 0.14; ...; 0.41	...	6206.............	0.43	...
			6234.............	0.02	...
5487.............	0.30; 0.11; ...; 0.250	...	6270[c]............	0.39; 0.12; 0.03; 0.30	...
5494[d]............	0.04	...	6284[c]............	2.00; 0.16; 0.10; 0.63	...
5508[d]............	0.03	5503 (w)			
5535[c]............	0.53; 0.19; ...; 0.46	...	6314.............	0.80	6314 (w)
			6354.............	0.06	...
5545[d]............	0.04	...	6363.............	0.04	...
... .	...	5658 (m)	6376[d]............	0.09; 0.04; 0.07; 0.11	6376 (w)
5705[c]............	0.29; 0.07; 0.06; 0.27	...	6379[d]............	0.16; 0.04; 0.05; 0.19	...
5778[c]............	0.95; 0.23; ...; 0.89	...	6426.............	0.04	...
5780[c]............	0.88; 0.24; 0.16; 0.80	5784 (m)	6445.............	0.09	...
			... .	...	6520 (s)
5795[c]............	0.39	...	6597.............	0.02	...
5797[d]............	0.39; 0.14; 0.08; 0.33	...	6614[d]............	0.40; 0.16; 0.14; 0.49	...
5844.............	0.14	...	6661.............	0.1; ...; 0.09; 0.08	...
5850[d]............	0.10; 0.07; 0.02; 0.13	5848 (w)			
			6699.............	0.06	...
... .	...	5965 (s)	6742.............	0.06	6741 (vs)
6011.............	0.19	...			
6042.............	0.31	...			

[a] From Herbig (1975, 1988); data correspond to observations toward HD 183143, HD 167264, HD 26571, and HD 168625, respectively
[b] Salama & Allamandola 1991a.
[c] Denotes Krelowski 1989 "5780 Å" family.
[d] Denotes Krelowski 1989 "5797 Å" family.

TABLE 2

NAPHTHALENE CATION VIBRONIC TRANSITION BAND WIDTHS, ABSORPTION COEFFICIENTS,
OSCILLATOR STRENGTHS, AND CROSS SECTIONS

λ (Å)	v (cm^{-1})	Δv (cm^{-1})	ϵ_{max}^{+} (l mol^{-1} cm^{-1})	f	σ_e (cm^2 molecule^{-1})
6741..................	14835	120	900	0.00040	3.4×10^{-18}
6520..................	15337	100	400	0.00010	1.5×10^{-18}
6151..................	16258	100	410	0.00010	1.6×10^{-18}
5965..................	16765	80	200	0.00005	7.6×10^{-19}
				($\Sigma = 0.00065$)	
4560..................	21930	140	43	0.00002	1.6×10^{-19}
3769..................	26532	400	100	0.00010	3.8×10^{-19}
3050..................	32787	215	1040	0.00080	4.0×10^{-18}

NOTE.—Neon matrix results from Salama & Allamandola 1991a.

sity mismatch between the $C_{10}H_8^+$ band intensities and the DIB equivalent widths measured by Herbig (1975) in the specific case of HD 183143. Moreover, with the remarkable exception of the very strong 6741 Å band, none of the stronger $C_{10}H_8^+$ bands have known DIB counterparts while the weaker members do show a surprising coincidence. However, one should keep in mind, when going through such comparisons, that the intensity ratios of the DIBs are highly variable from one object to another since the measurements often constitute an ill-defined average over all clouds situated along the line of sight (see Table 3 in Herbig 1975; Krelowski 1989). Hence, one can imagine that the weaker naphthalene bands might contribute to the DIBs and the strongest bands have not yet been recognized as DIBs. However, if this is the case, it should be established why the stronger $C_{10}H_8^+$ bands are not observed (i.e., if those features are hindered by stellar, atmospheric or instrumental features or if they simply escaped detection). Consequently, *observations should be carried out to search for the stronger bands of* $C_{10}H_8^+$—*which are predicted at 6520 Å, 6151 Å, and 5965 Å* (Salama & Allamandola 1991a)—providing a test for the presence of this specific ion in the interstellar medium. The small number of DIBs ($\sim 15\%$) which could be accounted for by $C_{10}H_8^+$ shows that $C_{10}H_8^+$ *alone cannot explain the DIBs in this wavelength range.* This comparison hints, however, that a family of different PAH ions may well explain some (or most?) of the DIBs. The answer to this puzzling problem can be obtained only through a detailed and systematic comparison of the DIBs with appropriate laboratory data.

The second approach would be to soften the requirements for a molecular species to contribute to the DIBs by requiring that only the strongest PAH ion band be evident and that subsequent searches be made for the less intense bands. If this criterion is adopted, the match between the strongest naphthalene cation band at 6741 Å and the 6742 Å DIB is intriguing. Using the $C_{10}H_8^+$ oscillator strengths summarized in Table 2, we calculate that 0.3% of the available carbon would be required to be in the form of $C_{10}H_8^+$ if the naphthalene cation is responsible for the 6742 Å DIB.

2. Another important result of these experiments is *the production of a broad and strong continuum extending from the UV to the visible* (2000–3800 Å with a weak tail extending to ≈ 5100 Å) which appears to be correlated with the discrete features associated with the cation. Figures 2 and 3 show the correlated growth of the cation discrete bands and continuous absorption with VUV irradiation time. These spectra also indicate that the growth of the cation continuum and discrete

absorption features by VUV irradiation is correlated to the depletion of the neutral precursor absorption.

Because of its potential astrophysical importance, the experiments described in Salama & Allamandola (1991a, b) were carried out to test such other possible continuum carriers as (1) the anionic counterpart ($C_{10}H_8^-$) which presumably forms in the matrix (by charge conservation), (2) a dissociative transition within the neutral precursor, (3) radicals produced by VUV irradiation, (4) a photochemically generated residue, (5) charge transfer with the matrix material induced by the high-energy (10.2 eV) photons, and (6) drift of the baseline due to a time-related drift in the response curve of the spectrometer.

The results of these experiments eliminated these possibilities and point to the positive naphthalene ion as the carrier of the continuum. As a further test of this conclusion, we have plotted in Figure 4 some representative absorbance values of the continuum together with the optical depth of some $C_{10}H_8^+$ discrete bands as function of VUV irradiation time. Figure 4 clearly shows that the discrete bands due to $C_{10}H_8^+$ and the continuum grow together. On the basis of this experimental study, we tentatively conclude that the continuum observed in our experiments in the 2000–5100 Å range is due to the naphthalene cation ($C_{10}H_8^+$). In the absence of any relevant theoretical calculations (as far as the authors know), it is premature to attempt a spectroscopic assignment of this transition.

Table 2 shows the oscillator strength (f) and molar absorption coefficients (ϵ) associated with the discrete $C_{10}H_8^+$ bands, while in Table 3, the continuum profile and strength is given in terms of the molar absorption coefficient (ϵ) versus the wavelength. At the peak, the cross section of the continuum is 7.6×10^{-15} cm^2 molecule^{-1}

TABLE 3

MOLAR ABSORPTION COEFFICIENTS AND CROSS SECTIONS FOR THE "2900 Å" CONTINUUM

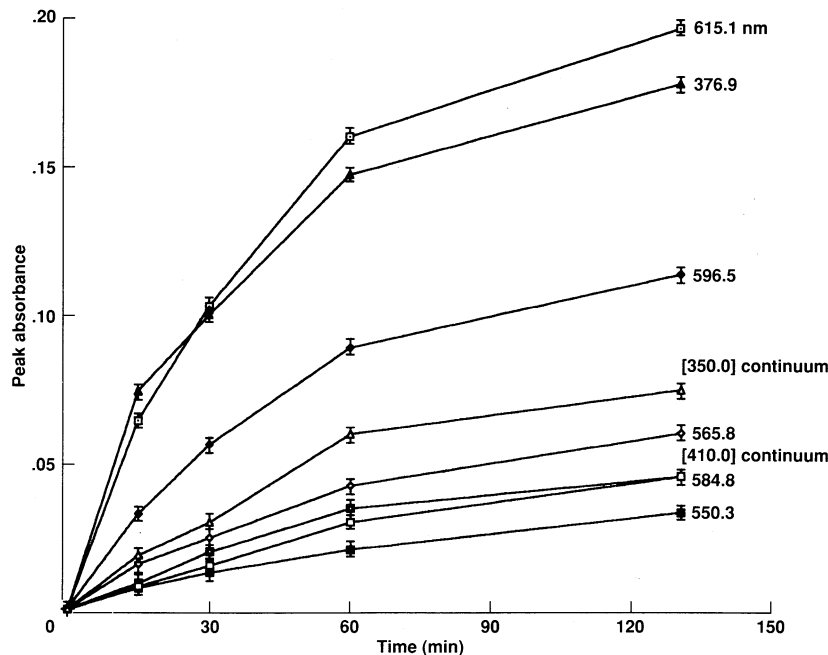| λ (Å)	σ_e (cm^2 molecule^{-1})	ϵ (l mol^{-1} cm^{-1})
2200...........	0	0
2400...........	2.3×10^{-15}	0.6×10^6
2600...........	6.1×10^{-15}	1.6×10^6
2800...........	7.6×10^{-15}	2.0×10^6
3000...........	7.6×10^{-15}	2.0×10^6
3200...........	5.0×10^{-15}	1.3×10^6
3400...........	5.0×10^{-15}	1.3×10^6
3600...........	3.8×10^{-15}	1.0×10^6

Fig. 4.—The correlated growth of the visible discrete absorption bands and continuum of $C_{10}H_8^+$. $Ne/C_{10}H_8 = 800$, $T = 4.2$ K.

If a broad UV–visible continuum is indeed an intrinsic spectroscopic property of the free, gas phase naphthalene (and other PAHs) cation and not a matrix effect, it is of great astrophysical significance. This strong continuum could provide *the channel by which interstellar UV–visible radiation is converted to the discrete IR emission bands in UV-poor regions.* The ability of the IR feature–emitting material to absorb in the visible is required to account for the IR emission intensities from several objects (Sellgren 1984; Sellgren et al. 1990; Aitken & Roche 1983). These structureless continua would blend, contributing to the visible portion of the interstellar extinction curve, but be largely undetectable, eliminating one of the criticisms of the PAH model—namely that band structure should be evident in the UV—(Donn, Allen, & Khanna 1989). (Note

that a mixture of a large number of neutral PAHs also washes out near VUV band structures [Léger et al. 1989]). They could also be responsible for the very broad structure (VBS) on the extinction curve (Hayes et al. 1973; van Breda & Whittet 1981). In view of this, it is extremely important to experimentally determine if this continuum is indeed a general property of ionized PAHs and if so, the nature of this transition.

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