Unidentified Cometary Emission Lines as the Photoluminescence of Frozen Hydrocarbon Particles

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Abstract—We discuss the possible nature of unidentified cometary emission lines. We propose a model of the ice particles in cometary halos as a mixture of frozen polycyclic aromatic hydrocarbons (PAHs) and acyclic hydrocarbons. We describe the general properties of frozen hydrocarbon particles (FHPs) and suggest interpreting some of the unidentified cometary emission lines as the photoluminescence of FHPs. We compare the positions of unidentified emission lines in the spectrum of Comet 122P/de Vico with the positions of quasi-lines in the photoluminescence spectrum of PAHs that were dissolved in acyclic hydrocarbons at a temperature of 77 K and that constituted a polycrystalline solution. We estimate the detectability of FHP photoluminescence in cometary spectra. © *2004 MAIK "Nauka/Interperiodica"*.

Key words: *Solar system—planets, comets, small bodies, heliosphere; unidentified cometary emission lines; frozen hydrocarbon particles; polycyclic and acyclic hydrocarbons; photoluminescence.*

INTRODUCTION

The problem of unidentifiable cometary emission lines in the optical and other parts of the cometary spectrum is known to remain of great interest. Emission lines of this kind defy any numerical or comparative identification and are generally collected in separate tables or lists (Brown *et al.* 1996; A. Cochran and W. Cochran 2002). Successful identification of these emission lines is associated with the upgrading of spectroscopic data acquisition and processing methods, the accumulation of laboratory comparative material, and the study of other excitation mechanisms for the corresponding emission lines. Of course, unidentified emission lines can arise within the framework of the standard resonance—fluorescence mechanism through transitions from higher levels of the corresponding atoms and molecules of cometary gases. However, the possibility that the photoluminescence of solid cometary matter is excited by solar ultraviolet radiation (Churyumov and Kleshchonok 1999; Simonia 1999) should be taken into account. The probability of this process is high, particularly if the presence of an appreciable amount of an organic component in cometary ices is taken into account. Polycyclic aromatic hydrocarbons (PAHs) are known to be among the most important classes of chemical compounds encountered in various bodies in space. Their presence in cometary matter is also beyond question (Ehrenfreund and Charnley 2000).

In our view, the ices of cometary nuclei can contain mixtures of PAHs and acyclic hydrocarbons. Consequently, the ice particles of the halo that surrounds a cometary nucleus can be composed entirely of these mixtures or contain appreciable amounts of them. Solar ultraviolet radiation will excite the photoluminescence of ice particles in the halo. The low albedo of the particles composed of these mixtures and the high quantum yields of photoluminescence for PAHs ensure the detectability of the corresponding luminescent emissions.

Under low temperatures, PAHs and acyclic hydrocarbons, as well as other possible components, will be in a frozen state. We propose calling the particles of this kind frozen hydrocarbon particles (FHPs). Let us consider the FHPs and their luminescence in more detail.

FHP LUMINESCENCE

Here, we first provide information about the quantum yields of luminescence for organic compounds. Witt and Vijh (2004) point out that the quantum yield of luminescence for many organic compounds is about 50%. They also distinguish PAHs from other materials by their highly efficient luminescence and note that, in certain cases, PAHs and their ions exposed to far ultraviolet radiation are capable of emitting several luminescent photons with a total quantum yield as high as 100% after a single excitation event.

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D'Hendecourt *et al.* (1986) noted that the quantum yield for PAH molecules could reach 50%.

Gudipati *et al.* (2003) believe that the quantum yield for small grains containing frozen organic mixtures can vary within the range 90−100%. The aforesaid suggests that PAHs have highly efficient, intense luminescence when exposed to ultraviolet radiation. Mixtures of frozen PAHs and acyclic hydrocarbons may be contained in the ices of cometary nuclei. We assume that these mixtures are solid solvent−material solutions where by the material and the solvent we mean PAHs and acyclic hydrocarbons, respectively. In other words, the PAHs dissolved in acyclic hydrocarbons constitute a single solid solution with it. The optical properties of the solution are determined by the properties of the solvent, the solution crystallization conditions, the presence of a luminescent component (luminogen), the pattern of interaction between the components of the solution, and the content of the admixtures in the solution.

The presence of polycrystalline solutions in cometary ices is quite probable. Patashnick *et al.* (1974) and Smoluchowski (1981) showed, in particular, that amorphous ice in comets transforms into a crystal with a cubic lattice at a temperature of about 140 K. The surface layers of an ice cometary nucleus are the sources of variously sized frozen hydrocarbon particles ejected and carried away into the circumnuclear region as the heliocentric distance of the comet decreases. These ice particles of various sizes and fragment symmetry form an ice halo at the corresponding heliocentric distances. Fernandes and Jockers (1983) believe that an ice cometary halo at a heliocentric distance of $r = 1$ AU can have a radius as large as 500 km. This value is debatable, particularly if the density of the solid particles under consideration is taken into account, but it is quite acceptable for a general order-of-magnitude estimate of the halo radius. At the same time, it should be noted that space missions to comets have not yet confirmed the presence of ice halos around them. Therefore, the existence of halos of icy particles around cometary nuclei is still open to question and is considered as a hypothesis. In our view, the FHP sizes can vary from microns to millimeters. FHPs will have the color characteristic of a frozen mixture of PAHs and acyclic hydrocarbons. For low concentrations of the admixtures, the FHPs will be gray ice particles. Thus, the cometary nuclei may be surrounded by halos in the form of FHP layers. Under actual conditions, apart from FHPs, the halos may contain silicate dust and fine carbonaceous particles. When exposed to solar ultraviolet radiation and solar wind particle streams, FHPs will intensely luminesce in the wavelength range 3800−6700 Å. Other constituents of the halo (e.g., inorganic dust grains) will also luminesce, but

with a lower quantum yield. The FHP luminescence spectrum will be determined by the specific chemical composition of the mixture, the PAH concentration in the polycrystalline solution, the presence or absence of admixtures, the particle temperature, the phase of solar activity, and other factors. The most important factors are the chemical composition of the specific FHP and its temperature. For chemically different FHPs with different temperatures, solar ultraviolet radiation will excite luminescence of different spectral composition. Pringsheim (1949) noted, in particular, that the diffuse spectral bands of many crystalline organic compounds at a liquid oxygen temperature of 54.3 K are resolvable into groups of narrow bands or lines. This is very important for FHPs and suggests that the ice particles of cometary halos may have luminescence spectra of at least two types: (1) spectra composed of broad diffuse bands and (2) spectra in the form of a series of narrow bands or lines. A close correlation between the types of spectra for actual FHPs and their temperatures can be found only experimentally by comparing observational and laboratory data.

Let us now describe some of the FHP properties in the temperature range 60−80 K. For this purpose, we here provide data on the laboratory FHP substance analogs. Teplitskaya *et al.* (1978) produced polycrystalline solutions of the above chemical composition to study the luminescence of PAHs at a temperature of 77 K. They showed that normal paraffins were convenient solvents for obtaining discrete PAH luminescence and absorption spectra. For compounds with a linear structure (polyacens, polyphenyls, diphenylpolyenes, etc.), the sharpest spectra are observed when the linear sizes of the solvent molecules are close to those of the PAH molecules.

Experiments indicate that highly structured hydrocarbon spectra with line widths within the range 0.15−0.47 Å can be obtained when aromatic hydrocarbon solutions are excited by laser light into the (0−0) band at $T = 4.2$ K. This is true for most of the crystallizable and vitrifiable (when frozen) solvents that are chemically neutral with respect to the molecules dissolved in them and optically transparent in the absorption and emission ranges of PAH molecules.

Experiments also indicate that the luminescence spectrum of the mixture in certain solutions at a low PAH concentration consists of a series of bands, but a series of lines can appear as the PAH concentration increases. The luminescence spectrum for pyrene with a concentration of 10^{-4} mol l^{-1} in n-pentane contains no narrow lines. Shpol'skiĭ (1962) showed that when PAHs were dissolved in special hydrocarbon (e.g., n-pentane) solvents at a temperature of 77 K or lower, a frozen mixture of these materials had a luminescence spectrum in the form of a series

of many narrow lines. Such spectra are called quasi-line luminescence spectra. The positions of these lines on the wavelength scale and their mutual arrangement and relative intensities are characteristic for each molecular structure. These data can be an indicator of a normal electronic–vibrational state of the corresponding molecular structures. We assume that the FHPs of cometary halos whose chemical composition is almost identical to that of the laboratory analogs can also have quasi-line luminescence spectra when exposed to ultraviolet radiation.

At the same time, it should be noted that the quasi-line luminescence spectra of FHPs under actual astrophysical conditions can differ slightly from laboratory ones, due to the peculiar temperature and pressure conditions in cometary comas, charged particle bombardment, and collisions with the gaseous neutrals of cometary atmospheres. The width of each line in the FHP luminescence spectrum must probably be a function of the heliocentric distance, because the temperature of the cometary matter varies with heliocentric distance. The lines in the quasi-line luminescence spectra of FHPs can broaden at temperatures $T > 80$ K and low PAH concentrations. A gradual broadening of the lines in the luminescence spectra of ice particles with decreasing heliocentric distance will be observed as the FHP volatile component is depleted. Thus, for example, if the FHP material is composed of PAHs and n-hexane, the depletion begins by the time an ice particle is heated to temperature $T \approx 290$ K at which n-hexane begins to evaporate intensely. Naturally, the luminescence spectrum of the corresponding FHP will change in this case. Thus, FHPs can be produced as sources of short-lived luminescence. Short-lived emission lines of this kind are commonly detected in cometary spectra. They are often unidentified. Thus, the pattern of the FHP luminescence spectrum for cometary halos must depend on the following factors: (1) the chemical composition of the frozen mixture, (2) the PAH concentration, (3) the heliocentric distance of the comet, and (4) the dynamical peculiarities of the halo. The role of the exciting radiation whose wavelength range and intensity will differ markedly from the laboratory ones should also be noted. The ice particles that constitute the halo will be exposed to short-wavelength solar radiation of a wide range and to solar wind charged particle streams.

The photoprocessing of FHPs by solar ultraviolet radiation will change the FHP luminescence spectrum both in composition and in intensity distribution of the luminescent emission lines in series. Note that these changes will take place during a certain period that may be designated as the FHP photoprocessing period. For each class of FHP, these periods at various heliocentric distances can vary between several hundred seconds and several years.

As we see, the picture is complex and changeable; it is determined by the ice particle temperature, the PAH concentration and composition, and the composition of the exciting radiation. However, for different comets at the same heliocentric distances (e.g., $r = 1$ AU), assuming the halo ice particles to be chemically identical, similar emission lines of a photoluminescent nature will be observed in the spectra of these comets. Thus, similar, previously unidentified emission lines can be observed in the spectra of different comets. We therefore suggest interpreting some of the unidentified emission lines in cometary spectra as the photoluminescence of FHPs. We calculated the ratio of the photoluminescence flux to the scattered solar radiation flux F_{lum}/F_{sc} for typical conditions of actual cometary halos composed of millimeter-size FHPs. The calculation was performed for the following conditions: the FHP is composed of phenanthrene ($C_{14}H_{10}$) + n-hexane at $T = 77$ K, and the phosphorescence line is $\lambda = 4602$ Å (Teplitskaya et al. 1978). We considered an FHP that scattered solar radiation with a wavelength of $\lambda = 4602.17$ Å.

According to A. Cochran and W. Cochran (2002), an unidentified emission line was observed in the spectrum of Comet 122P/de Vico at $\lambda = 4602.17$ Å.

We performed our calculations using Planck's formula and the expression for the energy exposure $E = \dfrac{(w/s)}{r^2}$ (I. Simonia and T. Simonia 2003) for $w = Ft$ (F is the flux, and t is the time), where w is the total energy of the radiation at the corresponding wavelength, s is the surface area of a halo with a radius of $R = 500$ km, and r is the heliocentric distance (1 AU). We assumed that the FHP luminescence yield was 50%, the FHP albedo was A = 0.1, and the wavelength of the exciting ultraviolet solar radiation was 2930.25 Å (the phenanthrene absorption range).

We obtained $F_{lum}/F_{sc} = 2.344$, implying that the luminescent signal lies above the scattered solar continuum as a weak but detectable emission line. Such relatively weak unidentified emission lines are widely encountered in the atlas by A. Cochran and W. Cochran (2002). The error within ±0.17 Å is small, particularly if the broadening of the corresponding lines described above is taken into account. The high quantum yield of luminescence for PAHs and the use of fast instruments and high-resolution spectrographs ensure that the luminescent signal is recognizable. We also calculated F_{lum}/F_{sc} for a different millimeter-size FHP: the FHP was composed of 1.2–5.6 dibenzanthracene ($C_{22}H_{14}$) + n-hexane at $T = 77$ K, while the phosphorescence

line was $\lambda = 6135$ Å (Teplitskaya *et al.* 1978); the FHP scattered solar radiation with a wavelength of $\lambda = 6135$ Å. According to A. Cochran and W. Cochran (2002), an unidentified line was observed in the spectrum of the same comet at $\lambda = 6135$ Å. We assumed that the FHP luminescence yield was 50%, the FHP albedo was A = 0.3, and the wavelength of the exciting solar radiation was $\lambda = 3660$ Å (the absorption range of 1.2–5.6 dibenzanthracene ($C_{22}H_{14}$)). Using the same formulas, we obtained $F_{\text{lum}}/F_{\text{sc}} = 1.552$. As we see, the luminescent signal also lies above the scattered solar continuum as a weak but detectable emission line.

COMPARATIVE ANALYSIS

We compared the laboratory luminescence spectra of FHP substance analogs with the observed cometary spectrum containing a set of unidentified emission lines. We used an atlas of quasi-line luminescence spectra for aromatic molecules (Teplitskaya *et al.* 1978) as the laboratory data and a high-resolution atlas for Comet 122P/de Vico (A. Cochran and W. Cochran 2002) as the observational data. The results obtained are summarized in the table[1]. It gives the PAH names and formulas (column 1), the solvent names (column 2), the wavelengths of the luminescent emission lines of the corresponding polycrystalline solutions (column 3), and the wavelengths of unidentified cometary emissions from the spectrum of Comet de Vico (column 4). We performed our comparative analysis with an accuracy of ± 1 Å.

DISCUSSION

Having described the concept of FHP and having performed a comparative analysis of the observational and laboratory data, the results of which are summarized in the table, we conclude that at least 28 aromatic hydrocarbons that constitute a polycrystalline solution with a number of acyclic hydrocarbons may be contained in the FHPs of the ice halo of Comet de Vico as the main mixture. Naturally, the FHPs will also contain admixtures in small amounts. In fact, the ice halo of Comet de Vico may be a complex of FHPs of various chemical compositions. A cloud of variously sized FHPs with different chemical compositions exposed to ultraviolet solar radiation will be a source of luminescent emission lines within a wide wavelength range, 3800–6700 Å. The laboratory comparison data used here covered both the fluorescence and phosphorescence of hydrocarbons.

The polycrystalline solutions, chemical FHP analogs, demonstrated intense fluorescence and phosphorescence under laboratory conditions.

In an actual space environment, the FHPs of cometary halos can have different afterglow periods for different excitation conditions; i.e., they can have fluorescence or phosphorescence spectra. The laboratory comparison database used here contained 100 aromatic molecules. The selection criterion that consisted in the match between the wavelengths of the corresponding emission lines to within ± 1 Å completely excluded 72 molecules and about 10% of the emission lines of the 28 selected aromatic molecules. The presence of PAHs in comets was discussed by a number of authors. In particular, Moreels *et al.* (1994) showed that phenanthrene exists in the inner coma of Comet P/Halley. This aromatic hydrocarbon was detected by the above authors in the near ultraviolet range of the cometary spectrum. The authors pointed out the stability of PAHs in a space environment and a possible connection between the PAHs contained in Solar-system bodies and interstellar PAHs. Crovisier and Bockelée-Morvan (1999) believe that the presence of PAHs in comets has not yet been conclusively proven. In their extensive review, Ehrenfreund and Charnley (2000) point out the detection of PAHs and aliphatic hydrocarbons in cometary matter.

Naturally, aromatic hydrocarbons can exist in comets in both condensed and gas phases. The described FHPs of the corresponding chemical composition are the carriers of a number of previously unidentified photoluminescent emission lines. At the same time, it is quite clear that all of the unidentified emission lines cannot arise from the photoluminescence of FHPs. A significant fraction of the unidentified emission lines (perhaps most of them) are of a resonance–fluorescence nature. The sources of these emission lines are the corresponding daughter molecules and ions. Newer comparative molecular emission line databases are required for their precise identification. Thus, only a small fraction of the unidentified cometary emission lines arise from the photoluminescence of FHPs. The nature of a large number of unidentified cometary emission lines remains unknown. It is highly likely that the gaseous components of cometary atmospheres are the sources of these emission lines (Wyckoff *et al.* 1994). When calculating the ratio $F_{\text{lum}}/F_{\text{sc}}$, we assumed the albedos of comets to be within the range 0.1–0.3. These values were suggested by Fernandes and Jockers (1983) and several other authors. Our calculations indicate that the luminescent signal will lie above the solar continuum at various albedos due to the high quantum yield of luminescence

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[1] The table is published in electronic form and is accessible via ftp cdsarc.u-strasbg.fr/pub/cats/J (130.79.128.5) or http://cdsweb.u-strasbg.fr/pub/cats/J.

Luminescence of cometary FHPs: The results of our comparative analysis

Column 1

PAH solvent	λ, Å	λ, Å
Pyrene C$_{16}$H$_{10}$		
n-Hexane	3929	3929.28
	5894	5893.85
	6027	6026.66
	6036	6036.42
	6346	6346.78
	6413	6413.07
	6509	6508.71
1,2−3,4 Dibenzanthracene C$_{22}$H$_{14}$		
n-Octane	3862	3862.07
	3936	3936.23
	3949	3949.15
	4069	4069.03
	4091	4090.99
n-Hexane	3034	3934.90
	3955	3955.13
	3981	3981.64
	4055	4054.64
	4067	4067.35
Pycene C$_{22}$H$_{14}$		
n-Hexane	3863	3863.16
	3871	3871.63
	3989	3989.37
	3999	3999.89
	4186	4186.75
1,2 Benzpyrene C$_{20}$H$_{12}$		
n-Hexane	3862	3862.07
	3960	3960.05
	3959	3959.12
	4074	4073.30
	4088	4087.97
	4103	4102.98
	4195	4194.46
	4212	4212.69
	4310	4310.10
	4324	4324.09
	5366	5366.02
	5349	5349.22
	5479	5479.25
	5718	5718.11
	5754	5753.51
	6016	6015.19
	6351	6351.65

Column 2

PAH solvent	λ, Å	λ, Å
Anthracene C$_{14}$H$_{10}$		
n-Hexane	3950	3950.02
	3963	3963.11
	3985	3985.44
	4011	4011.44
	4023	4023.07
	4075	4075.98
	4248	4248.06
	4264	4263.07
	4292	4291.73
	4490	4489.90
	4519	4519.09
n-Heptane	3867	3867.16
	3967	3967.51
	3987	3987.26
	4002	4001.97
	4025	4024.93
	4064	4064.46
	4094	4093.94
	4295	4294.88
	4541	4540.82
	4573	4572.97
Tetraphene C$_{18}$H$_{12}$		
n-Hexane	3884	3884.72
	3886	3886.61
	3934	3934.90
	3956	3956.48
	3961	3960.05
	3963	3963.11
	3999	3999.89
	4034	4034.01
	4044	4044.38
	4047	4046.74
	4063	4062.87
	4085	4084.85
	4096	4095.93
	6415	6415.14
	6439	6438.24
	6510	6509.91
	6638	6638.21
n-Octane	3891	3891.48
	3909	3908.67
	3912	3912.14

Column 3

PAH solvent	λ, Å	λ, Å
	3925	3924.82
	3929	3929.28
	4005	4005.94
	4038	4037.93
	4041	4041.94
	4046	4046.36
	4069	4069.03
	4070	4069.97
	4095	4094.78
	4098	4098.31
	5910	5909.17
	5935	5935.42
1,2−5,6 Dibenzanthracene C$_{22}$H$_{14}$		
n-Hexane	3936	3936.23
	3950	3950.02
	3953	3953.19
	4044	4044.38
	4045	4045.35
	4148	4148.93
	4195	4194.46
	5542	5542.40
	5942	5942.25
	6013	6012.37
	6096	6096.48
	6135	6135.00
1,2−4,5 Dibenzpyrene C$_{24}$H$_{14}$		
n-Hexane	3936	3936.23
	3950	3950.02
	3953	3953.19
	4002	4001.97
	4062	4062.86
	4186	4186.75
	4621	4620.97
	4657	4656.89
	4696	4695.85
	4809	4809.87
	4888	4888.45
Diphenil C$_{12}$H$_{10}$		
n-Hexane	4371	4371.16
	4570	4570.12
Fluoren C$_{13}$H$_{10}$		
n-Hexane	4239	4238.71
	4525	4525.13
	4552	4552.25

for PAHs. The use of fast instruments and high-resolution spectrographs ensures the detectability of FHP photoluminescence. In fact, A. Cochran and W. Cochran (2002) and Brown *et al.* (1996) were able to detect this photoluminescence.

Caution should be exercised in interpreting unidentified cometary emission lines as the photoluminescence of FHPs, because the quasi-line spectra of polycrystalline solutions have a peculiar feature: in several cases, a group of lines (multiplets) that are often repeated over the entire spectrum corresponds to each electronic—vibrational transition in the spectrum. The existence of several types of emitting centers in different local conditions, which causes the electronic levels to shift while the positions of the vibrational sublevels remain unchanged, may be responsible for the emergence of multiplets. In addition, some of the multiplet components may be associated with the presence of closely spaced levels for the same emitting center. The pattern of the multiplet also depends on the concentration of the dissolved material, the presence of admixtures, and the solution temperature and crystallization conditions.

Here, we have not considered the formation of a frozen mixture of PAHs and acyclic hydrocarbons. We intend to devote a separate paper to this subject. It is also important to consider the bombardment of FHPs by charged particle streams. Undoubtedly, the interaction of particles with cometary FHPs will have a serious effect on the pattern of their luminescence spectrum. We also intend to adapt the concept of FHP to the interstellar medium, i.e., to consider FHPs as the particles contained in such objects as reflection nebulae, complexes of gas and dust, etc.

CONCLUSIONS

We have presented the concept of FHP and described the salient features of their photoluminescence. We suggest interpreting at least some of the unidentified cometary emission lines as the photoluminescence of FHPs. We compared the spectra of laboratory FHP analogs with the observed spectrum of Comet de Vico containing several thousand unknown cometary emission lines. The results of our comparative analysis indicate that at least 28 aromatic molecules may be contained in the ice particles of the halo of this comet.

The results of our comparative analysis are summarized in the table. In our view, it would be appropriate to devote a series of separate laboratory studies to the modeling of FHPs and the excitation of their photoluminescence and to compile a database of unidentified cometary emission lines using several atlases and catalogs of cometary spectra. We realize that this paper is only the first step in this direction, and we will attempt to develop it by continuously upgrading our methods and improving our results.

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