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HYDROGENATED AMORPHOUS CARBONS PHOTOLUMINESCENCE AND ASTROPHYSICAL IMPLICATIONS FOR THE EXTENDED RED EMISSION

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Abstract. Hydrogenated amorphous carbons (a-C:H) have proved to be excellent analogs of interstellar dust through IR vibrational absorption bands (3.4 μm, 6.8 μm and 7.2 μm bands (respectively 2960 cm^{-1}, 1460 cm^{-1} and 1380 cm^{-1})) widely observed in galaxies diffuse interstellar medium (ISM). a-C:H are candidates for one of the observed interstellar dust features : a large emission band in the red part of the visible spectrum, attributed to photoluminescence (PL) of interstellar dust, and called the extended red emission (ERE). The PL absolute quantum yield is one of the strongest constraints set by such ERE observations. The PL relative quantum yield is known and measured for many a-C:H at discrete excitation wavelengths. The few absolute efficiencies determined are scattered and sometimes vary by orders of magnitude for supposedly identical a-C:H. We thus produce astrophysical a-C:H and analyze their PL and IR behavior, carefully accounting for thin film optical effects. By properly determining the excitation wavelength dependent PL absolute quantum yields for a wide variety of astrophysically relevant a-C:H, we can constrain these interstellar dust analogs as possible ERE candidates.

1 Introduction

The spaces between stars in galaxies are not empty but are filled with gas and dust which form the interstellar medium (ISM). The ISM permanently interacts with stars from their birth to their death. To understand this dynamical and chemical evolution, it is necessary to identify the ISM components. One of the goals of the interstellar medium research is to attribute the observed spectral features to carriers since most of accessible information about ISM results of its interaction with light. One of the ISM spectral features is the extended red emission (ERE): this is a large (FWHM between 60 and 120 nm) featureless emission band in the red part of the spectrum (between 540 and 950 nm) observed in many interstellar environments, first observed in the Red Rectangle nebulae by Cohen et al. (1975). An example of ERE spectrum, observed in the Red Rectangle nebulae (Schmidt et al. 1980) is represented on figure 3 (left). This spectral feature is probably due to interstellar photoluminescence (PL) initiated by absorption of UV-visible photons, but the identification of the ERE carriers remains an important issue. Many candidates have been proposed over the past decades.

Amorphous hydrogenated carbons (a-C:H or HAC) are one of the ERE carrier candidates. They are organic material of astrophysical interest since they have proved to be analogs of one of the significant interstellar dust components (Sandford et al. 1991; Pendleton & Allamandola 2002; Spoon et al. 2004; Dartois et al. 2005) through IR absorption bands (C-H stretching at 3.4 μm and C-H bending at 6.85 and 7.25 μm) ubiquitously observed in the diffuse interstellar medium of our Galaxy but also of other galaxies.

Amorphous hydrogenated carbons photoluminesce in the visible spectral range. It is thus important to characterize the photoluminescence behavior of this interstellar component that will contribute to the interstellar dust emission and compare the a-C:H PL to the ERE observed properties. In particular, the PL efficiency, which is for the ERE carrier candidates one of the strongest constraints set by the observations, has to be accurately investigated for a-C:H materials : until now, most of the works provided results in terms of relative efficiencies (e.g. Rusli et al. 1996). The results dedicated to astrophysical analogs about absolute a-C:H PL efficiencies (Furton & Witt 1993; Furton et al. 1999) vary by one order of magnitude, although the material is supposed to be similar and are determined with a simple optical description of what happens to the absorbed or emitted light in the sample. It is important to carefully study PL absolute efficiency of amorphous hydrogenated carbons, taking care to account for optical effects occurring in the sample bulk and at the thin film interfaces.

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2 Experimental Methods

We produced interstellar a-C:H analogs, using a plasma-enhanced chemical vapour deposition system with different hydrocarbons precursor gas, as films of few micrometers deposited on a substrate. We record ex-situ their IR and UV-visible transmission spectra allowing us to characterize them and to determine the parameters necessary to a complete description of their PL (optical band gap, absorption coefficient, film thickness).

The photoluminescence of the a-C:H films is recorded between 250 nm and 800 nm with a luminescence spectrometer, while the samples monochromatic excitation scan is selected in the UV-visible wavelengths greater than 250 nm. The intrinsic (internal) photoluminescence quantum yield, defined as the ratio of emitted to absorbed photons numbers, is calculated from the measured external PL by taking into account several effects due to the absorption and the interfaces of the a-C:H film. We developed an optical model based on previous work (Lukosz 1981; Holm et al. 1982; Nollau et al. 2000) that considers the thin film as an absorbing layer located between two different media and viewing the luminescent centers as electric dipole sources. Such an approach is required to determine the a-C:H intrinsic and absolute PL yield. This model includes the effective absorbed excitation quantity, the a-C:H film self-absorption of the PL emitted, the wide-angle and multiple reflections interference effects, the detection solid angle and the transmittance at the film-air interface, ... The PL efficiency determined is the intrinsic absolute PL quantum yield of the a-C:H material. It is important to obtain the intrinsic efficiency and not the external efficiency since the geometry of our samples is different from the interstellar grains one.

3 Results and Discussion

We produced and analyzed many different a-C:H samples covering an optical gap range E_{04} varying typically between 3.0 and 4.2 eV depending on the plasma production conditions. These few micrometers, transparent to yellow orange films are photoluminescent in the visible when illuminated by UV photons (when seen under a UV lamp, the a-C:H PL appears luminescent with a color varying from yellow to red). The wavelength of the photoluminescence maximum varies from about 460 nm to 650 nm as the optical gap energy decreases (see Fig. 1). As observed for the extended red emission (Darbon et al. 1999), the photoluminescence band FWHM is correlated to the variation of the band position : the band width increases with the band central wavelength for both ERE and a-C:H PL (see Fig. 2). However, the ERE width varies from 60 to 120 nm while the a-C:H PL we measured exhibits band widths between 130 and 340 nm, i.e about twice wider than for the ERE observations.

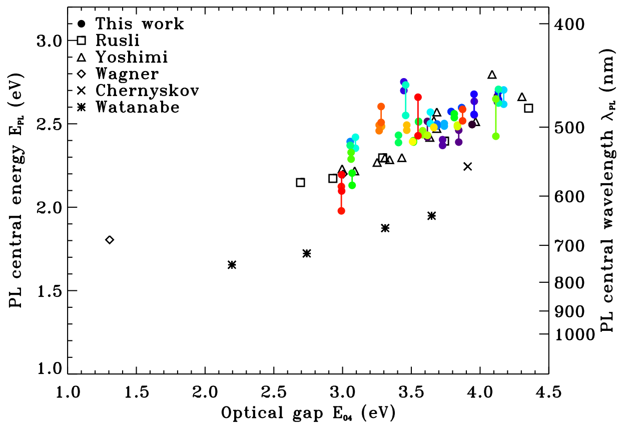

Fig. 1. Variation of the a-C:H photoluminescence color with the optical gap E_{04}. Other results are also plotted (Rusli et al. 1996; Yoshimi et al. 1992; Wagner et al. 1986; Chernyshov et al. 1991; Watanabe et al. 1982).

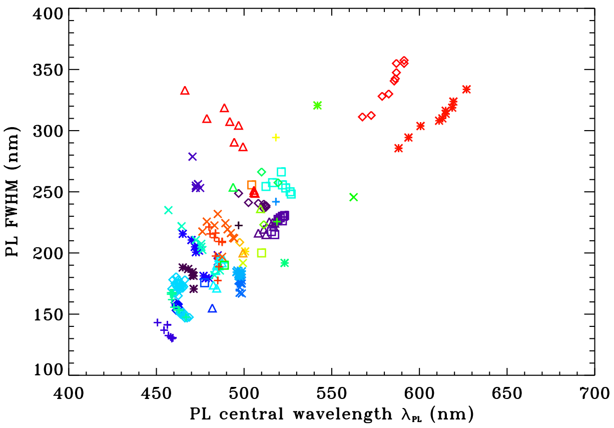

Fig. 2. Correlation of the FWHM with the central wavelength for the a-C:H photoluminescence bands. Different a-C:H samples produced are represented with di?erent symbols and colors.

To better correspond to the ERE spectra, the produced a-C:H should explore slightly lower optical band gaps than those we produce and have lower band width, in order to better overlap with the observational range. The figure 3 shows a comparison between a spectrum of the a-C:H PL and an ERE observation.

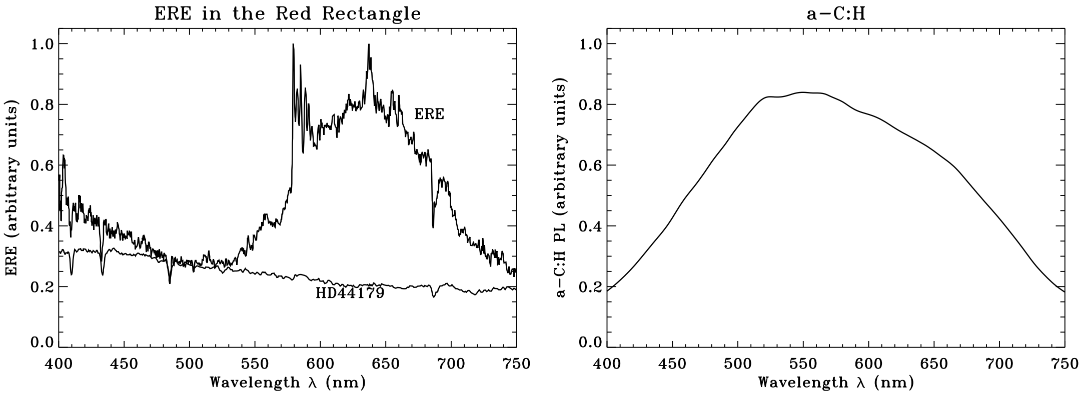

Fig. 3. *Left* : ERE spectrum observed in the Red Rectangle nebulae (and a reference spectrum of the Red Rectangle central star HD 44179) (Schmidt et al. 1980). *Right* : typical a-C:H photoluminescence spectrum.

We studied the influence of the excitation wavelength that gives rise to the photoluminescence : by varying this excitation between 250 and 490 nm, no strong modification of the photoluminescence band (position and width) has been observed and the PL quantum yield does not change by more than a factor of 2-3 (see Fig. 4).

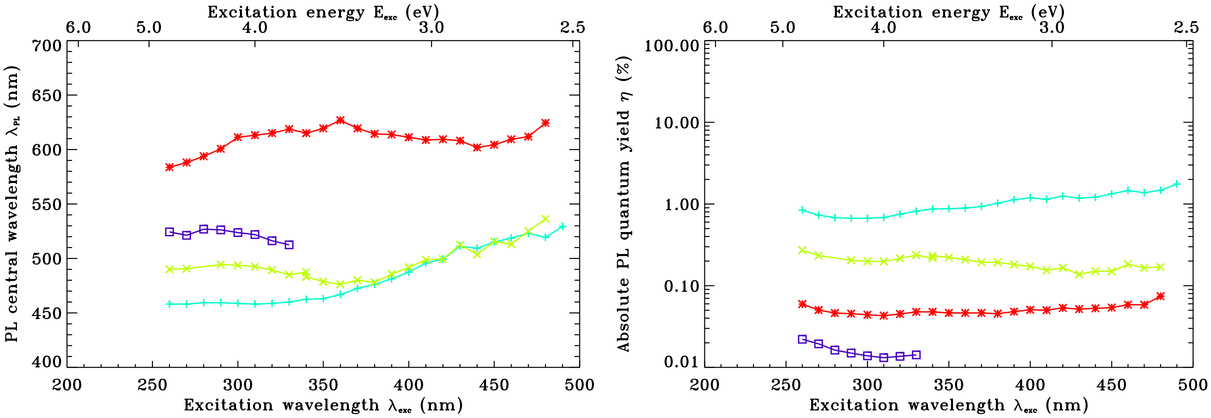

Fig. 4. Influence of the excitation wavelength on the a-C:H photoluminescence central wavelength (left) and efficiency (right) for four different a-C:H samples. By varying this excitation between 250 and 490 nm, no strong modification of the photoluminescence is observed (except the expected increase of the photoluminescence central wavelength when the excitation occurs in the PL band).)

As already shown in previous a-C:H photoluminescence studies (Rusli et al. 1996), we observed that the a-C:H PL yield decreases with the optical gap of the samples (see Fig. 5). Our work provides absolute PL yields ranging from few hundredths of percent to few percents in the optical band gap explored range. The astrophysical extended red emission observations provide lower limits for the carrier quantum yield, varying from one ISM environment to another. Near strong UV-visible stellar sources, the ERE efficiency lower limits are found to be the lowest ones (∼0.01%), and in the diffuse interstellar medium (DISM), high quantum yields lower limits of about 10% are estimated (Smith & Witt 2002). The corresponding yields correlate and increase with the decreasing density of the local radiation field. The quantum yields that we deduce are compatible with most of quantum yield limits set by ERE observations. However, they are not compatible to the highest quantum yield of 10% set by the scarce and difficult observations of the weakly emitting DISM.

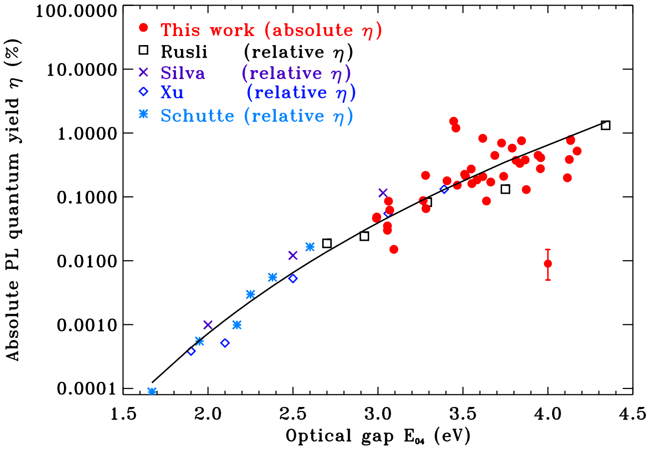

Fig. 5. Variation of the a-C:H photoluminescence quantum yield with the optical gap E_{04}. Typical error bar on the absolute yield including systematic effects are represented. Relative quantum yield results from other photoluminescence studies (Rusli et al. 1996 and references therein) are converted into absolute quantum yield using our absolute results and added to this figure. It allows to see variation of a-C:H PL absolute quantum yield on a larger optical gap range than with only our samples. The line is a fit to the relative efficiency data.

4 Conclusion

The evaluation of the astrophysical ERE efficiency from observations of different ISM environments, and more specifically the diffuse low density medium, is generally not a trivial task. The absolute yield, together with the elemental cosmic abundances requirements, are by far stronger constraints than the profile of astrophysical PL spectrum, as many materials are able to photoluminesce in the visible. Being the strongest astrophysical constraint, the PL efficiency determination need to be improved, in particular by insisting on additional observations of the extended red emission in the diffuse interstellar medium and proper evaluations of the UV-visible photons absorbed by the ISM carriers.

We provide in our study the first measurements of absolute PL yield as a function of excitation wavelength for a-C :H laboratory analogs to interstellar dust and a law linking the a-C:H optical gap to their absolute PL quantum yield comes out.

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