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UV photodestruction of CH bonds and the evolution of the 3.4 μ m feature carrier

II. The case of hydrogenated carbon grains

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Abstract. We present the results of a laboratory program aimed at studying the effects induced by energetic UV photons on hydrogenated carbon particles. Experiments have been performed under simulated diffuse and dense interstellar medium conditions. To monitor the effects of UV irradiation on grains IR spectroscopy has been used. In both circumstances UV photons lead to a reduction of the aliphatic 3.4 μ m band. An estimation of the destruction cross section by UV photons for the hydrogenated carbon particles has been derived from the reduction of the 3.4 μ m intensity band as a function of the UV fluence. The results of the present work, together with previous laboratory data, can shed light on the enigmatic difference observed for the 3.4 μ m band between dense and diffuse interstellar medium clouds. This difference is compatible with the transformation of hydrogenated carbon particles produced by UV photons and hydrogen atoms and with the changes of the grain properties in the two environments.

Key words. ISM: dust, extinction - infrared: ISM: lines and bands - methods: laboratory

1. Introduction

It is well established that carbonaceous materials are a primary component of the interstellar dust Henning & Salama 1998; Greenberg & Li 1999; (see Ehrenfreund & Charnley 2000 for recent reviews). Although current interstellar extinction models describe the carbon components and the physical link with silicate grains differently, their common aspect is the requirement of a considerable amount of the cosmic carbon abundance locked in dust (e.g. Mathis 1996; Li & Greenberg 1997). Specific spectroscopic features provide information on the composition of interstellar carbon materials, indicating the presence of both aromatic and aliphatic components in the interstellar medium.

The strongest interstellar dust feature, the UV extinction bump at 217.5 nm, is, indeed, clear evidence for a carbonaceous component with an aromatic character. Among the many proposed carbonaceous materials, graphite has long been considered a prominent candidate as the

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feature (e.g. Draine & Lee 1984; carrier of $_{\mathrm{this}}$ Mathis 1994). The graphite hypothesis has been questioned due to its shortcomings, which include, above all, the impossibility to reconcile it with the observed constancy of the peak position associated to the bump width variations (Greenberg & Chlewicki 1983; Draine & Malhotra 1993). It is now generally accepted that nanosized carbon grains play a fundamental role as carrier of the UV bump. This interpretation is supported by a number of experimental and theoretical studies (Mennella et al. 1996a, 1997, 1998; Duley & Seahra 1998; Schnaiter et al. 1998). A good understanding of the reasons for the observed bump profile variations has recently been obtained in terms of the contribution of different nanosized hydrogenated carbon grains with different degrees of UV processing along different lines of sight (Mennella et al. 1998).

In addition to the UV bump, which can provide clues to the electronic structure of carbon dust, information on its vibrational properties is inferred from the study of spectral IR signatures. Infrared spectra of lines of sight containing dust in the diffuse interstellar medium show the presence of an absorption band at 3.4 μ m (Adamson et al. 1990; Sandford et al. 1991; Pendleton et al. 1994). This feature is characteristic of the CH stretching modes in the methyl (CH_3) and methylene (CH_2) groups of aliphatic compounds. The 3.4 μm band has also been detected in the spectrum of the C-rich protoplanetary nebula CRL 618 (Lequeux & Jourdain de Muizon 1990; Chiar et al. 1998). The remarkable similarity of the band profile to that of the Galactic center indicates that the carriers of the interstellar band may have a partly circumstellar origin (Chiar et al. 1998). There is, however, a distinct absence of the 3.4 μ m absorption band in the spectra of molecular cloud dust; in this case a feature at 3.47 μ m characterises the CH stretching region (Allamandola et al. 1993; Chiar et al. 1996; Brooke et al. 1996).

To get constraints on the nature of the aliphatic component, Pendleton et al. (1994) have performed spectral comparisons of the feature with that of analogue materials such as refractory carbonaceous residues formed by the warming of UV photolyzed and ion bombarded interstellar ice analogs, quenched carbonaceous composites, hydrogenated carbon grains and a carbonaceous fraction of the primitive meteorite Murchinson. Greenberg et al. (1995) have obtained a good fit to the feature with laboratory residues of photo-processed ices which were exposed to solar UV radiation on the EURECA satellite. Recent mass spectroscopy measurements have indicated that these photo-processed residues contain a rich mixture of polycyclic aromatic hydrocarbons (Greenberg et al. 2000). These comparisons indicate that materials that contain substantial amounts of aliphatic CH_2 and CH_3 groups can reproduce the interstellar feature with varying degrees of accuracy. The main difficulty for a unique identification of the feature carrier is that IR spectroscopy gives information on the local bonding of the CH bonds and the band profile is rather insensitive to the details of the carbon structure (e.g. Sandford et al. 1991; Pendleton et al. 1994; Furton et al. 1999).

Information on the composition of the material responsible for the aliphatic feature may be obtained by analyzing the differences between dense and diffuse environments. In fact, in view of the continuous cycling of material between dense and diffuse phases of the interstellar medium (e.g. Greenberg & Li 1999), the observed difference represents a strong constraint for any description of the carrier. The effects of two competitive processes, the formation of CH bonds by H atoms interacting with carbon based materials and their destruction by UV photons, should be taken into account for modelling the 3.4 μ m band evolution. Of course, the efficiency of these interactions under dense and diffuse interstellar medium conditions has to be determined carefully.

In this context, the study in the laboratory of the transformations induced in carbonaceous materials by UV photons and H atoms is fundamental to get more insight into the evolution of the 3.4 μ m band carriers in

different environments. Recently, Mennella et al. (1999) have shown that hydrogenation by H atoms of hydrogenfree nano-sized carbon grains takes place under simulated diffuse interstellar medium conditions. On the other hand, it is known that UV irradiation decreases the intensity of the CH stretching feature in hydrogenated carbon materials (Iida et al. 1984). This trend is confirmed by the systematic study on several hydrocarbon molecules reported in Muñoz Caro et al. (2001), hearinafter Paper I.

In this paper we report on new laboratory results aimed at studying the evolution of the 3.4 μ m band carriers in different environments. The main aim of the present work is to evaluate the UV photo-destruction cross section for hydrogenated amorphous carbon grains and to extend the analysis of Paper I. In Sect. 2 we briefly describe the set-up adopted to produce and irradiate grains and we present the results. They are discussed in Sect. 3 while the astrophysical implications of the experiments are reported in Sect. 4. Finally, the main conclusions of the work are summarized in Sect. 5.

2. Experiment and results

The hydrogenated carbon grains (hereinafter ACH2) studied in this work were prepared by condensation of carbon vapour obtained by striking an arc discharge between two carbon rods in a 10 mbar hydrogen atmosphere. The carbon particles were collected on CsI substrates located 5 cm from the source. The resulting samples are characterized by predominantly chain-like aggregates composed of spherical grains with an average diameter of 10 nm. Rare forms of poorly graphitized carbon, bucky-structures and graphitic structures are also observed (Rotundi et al. 1998). The optical properties of these carbon grains have been studied in previous work to which we refer the reader for more details (Colangeli et al. 1995; Mennella et al. 1995). ACH2 was chosen because it can be considered a good analog of the 3.4 μ m band interstellar carrier. Its aliphatic CH stretching mode at 3.4 μ m is much stronger than the aromatic feature near 3.3 μ m and the band profile is similar to that observed toward the Galactic center source IRS 6E by Pendleton et al. (1994) (see Fig. 1).

To study the effects induced by UV photons in hydrogenated carbon grains, ACH2 samples were irradiated, at low temperature (~12 K) and at a pressure of 10^{-7} mbar by using a microwave excited hydrogen flow discharge lamp with a MgF₂ window. At the sample position the source provides a flux of UV photons, with an energy ≥ 6 eV, of ~5 10^{14} photons cm⁻² s⁻¹. Further details of the experimental set-up are reported in Gerakines et al. (1995) and in Paper I.

The irradiation experiments were aimed at simulating UV processing of grains under dense and diffuse medium conditions. In the first case, to reproduce the icy mantle accreting on grains, a water ice layer was deposited on ACH2 grains. The ice mixture $H_2O:CO:NH_3 = 100:33:16$ was considered for comparison, to see whether the

Table 1. UV irradiation experiments of ACH2

No.	Ice deposit	$d(ice)^a$ μm	$F_{\max}^{\ b}$ 10 ¹⁹ photons cm ⁻²	$ au_0^{\prime \ c}$	$ au_1^{\ c}$	$\sigma_{\rm des}{}^c$ $10^{-19} {\rm cm}^2/{\rm photon}$
$\frac{1}{2}$	H ₂ O H ₂ O:CO:NH ₃ 100:22:16	0.019 0.011	1.7 2.8	0.47 ± 0.05 0.62 ± 0.06	0.24 ± 0.05 0.19 ± 0.06	1.1 ± 0.3 0.8 ± 0.2
3	Ar	~ 0.5	2.7	0.41 ± 0.04	0.47 ± 0.04	1.0 ± 0.3

a) Ice thickness.

b) Maximum UV fluence.

c) Best fit parameters of Eq. (2) to the data reported in Fig. 3.



Fig. 1. The 3.4 μ m band of ACH2 compared with the interstellar feature towards the Galactic center source IRS 6E. Open and filled circles refer to high and low resolution observations, respectively (Pendleton et al. 1994)

chemical composition of the ice can affect the processing. To emulate the diffuse interstellar medium conditions, the carbon grains were covered with a layer of argon to prevent any direct contact by contaminant background gas in the vacuum chamber (Gerakines et al. 1996). The thickness, d(ice) of the ice layer for the three experiments is reported in Table 1.

The IR spectral evolution of the samples was monitored during UV irradiation by recording periodically the spectrum at a resolution of 2 cm⁻¹ with a FTIR spectrophotometer. The behaviour of the 3.4 μ m band at low temperature before irradiation and after the maximum UV processing is shown in Fig. 2 for the three experiments. As one can see UV processing produces a significant decrease of the feature. The evolution of the integrated intensity of the 3.4 μ m band, τ , as a function of the UV



Fig. 2. The 3.4 μ m band at low temperature after the ice deposition (continuous line) and after the maximum irradiation (dashed line): ACH2 + H₂O ice layer a), ACH2 + H₂O:CO:NH₃ = 100:33:16 ice mixture b) and ACH2 + Ar ice deposit c)

fluence obtained in the three experiments is reported in Fig. 3: a systematic decrease of τ with UV irradiation is observed. At the end of irradiation, after warming up of the samples to evaporate the ice layer, the residual band is still present and ACH2 is not destroyed.

We note that water accretion on the sample due to imperfect vacuum conditions takes place during the long exposures we considered. This deposit absorbs part of the photons impinging on the sample and, consequently, reduces the actual fluences. We have estimated the reduction by evaluating the column density of the accreted water molecules from the increase of the 3 μ m band by using a UV cross section of 2 10⁻¹⁸ cm² per molecule (Okabe 1978). The fluences of Fig. 3 have been corrected for this effect. The maximum fluence, $F_{\rm max}$, for the present experiments is listed in Table 1. Quantitative information on the efficiency of CH bond photodestruction can be obtained from the observed trend. An exponential decay with the irradiation time t is expected for τ (see Paper I):

$$\tau = \tau_0 \, \exp -(\phi_{\rm UV} \, \sigma_{\rm des} \, t). \tag{1}$$

Here, τ_0 is the band intensity before irradiation, $\phi_{\rm UV}$ the UV flux at the sample position, σ_{des} is the destruction cross section of CH bonds per UV photon and t is the time of irradiation. This relation is based on the assumption that the efficiency of the CH bond photodissociation is constant and that the sample processing is uniform, since τ goes to zero for long exposures, after all the aliphatic component is destroyed. Moreover, to interpret the reduction of the band intensity as a reduction of the number of CH bonds during irradiation, the sample has to be optically thin at 3.4 μ m. The CH stretching feature of the ACH2 samples studied in the present work is indeed optically thin (see Fig. 2), however, at UV wavelengths the samples are optically thick as can be concluded from the band optical depth and from the absorption coefficient of ACH2 reported in Colangeli et al. (1995). UV processing is not uniform and the deeper grain layers are less processed than top layers. A similar situation should not occur for interstellar carbonaceous materials. In that case processing should be uniform since the characteristic thicknesses of interstellar materials are optically thin at far UV wavelengths.

To take into account the non uniform processing of our samples we slightly modified the relation (1) as follows:

$$\tau = \tau_1 + \tau'_0 \exp{-(\phi_{\rm UV} \ \sigma_{\rm des} \ t)}.$$
 (2)

The aymptotic value τ_1 represents the residual 3.4 μ m band intensity for long UV irradiation times, due to the unprocessed deeper layers of the samples. $\tau'_0 + \tau_1$ is the starting band intensity and τ'_0 is the maximum reduction expected for the band intensity. The best fits of relation (2) to the experimental data are shown in Fig. 3, while the best fit parameters are listed in Table 1.

The destruction cross sections are equal within the errors for the experiments simulating grain processing under dense medium conditions. However, the cross sections refer to samples with different ice thicknesses. To be accurate one should compare the results obtained for similar ice deposits. To do this we assume as a reference the ice thickness of ~0.01 μ m (experiment 2) and correct the cross section of the experiment 1 for the absorption of the UV flux due to the ice deposit in excess relative to the reference value. The correction corresponds to a contraction by a factor 1.05 of the UV fluences, which implies an increase of the same factor for σ_{des} . Our previous conclusion remains valid due to the small correction factor.

The estimated destruction cross section for experiment 3 falls between the values obtained for the other experiments. Therefore, an average value of $(1.0 \pm 0.2) \ 10^{-19} \ \text{cm}^2/\text{photon}$ can represent the photodescruction cross section of hydrogenated carbon grains under



Fig. 3. Evolution of the 3.4 μ m band intensity with UV irradiation for ACH2 + H₂O ice layer a), ACH2 + H₂O:CO:NH₃ = 100:33:16 ice mixture b) and ACH2 + Ar ice deposit c)

simulated diffuse and dense medium conditions. From the estimated parameters we obtain a reduction of 94% of the 3.4 μ m band intensity for the maximum fluence of 2.8 10¹⁹ photons cm⁻² (see Table 1). The aliphatic CH bonds are almost completely destroyed in the processed layers of our samples.

The present results indicate a lower destruction cross section for hydrogenated carbon grains than for hydrocarbon molecules (see Paper I). This difference is in line with a decrease of the CH destruction efficiency by UV photons found in Paper I for aliphatic and aromatic hydrocarbon molecules. In fact, the aromatic clustering degree of the hydrogenated carbon particles we have considered is substantially higher than that of a molecule such as ethylbenzene. This structural difference can lower the destruction cross section as discussed in Paper I. Moreover, UV irradiation measurements of hydrogenated amorphous carbon films (a–C:H) have been performed by Iida et al. (1984). They observed a film photodarkening (i.e. a change of the absorption edge, which corresponds to a decrease of the optical gap from 2.2 to 2 eV) and a decrease of the 3.4 μ m band peak intensity after irradiation with 3.4 eV photons. From the data they report we estimate a photodestruction cross section of $1.2 \ 10^{-20} \ \mathrm{cm}^2/\mathrm{photon}$, lower than the value obtained for the present experiments. In this case, in addition to structural differences between the two materials, the photon energy used in the two experiments $(\sim 10 \text{ vs. } 3.4 \text{ eV})$ can play a fundamental role to explain the different results. Finally, Ogmen & Duley (1988) did not mention variations of the 3.4 μ m in their a–C:H film after exposure for several hours to the full light from a

deuterium lamp. In this case a quantitative comparison with our results is not possible, since they did not report the fluence for their irradiation.

3. Discussion

The present understanding of hydrogenated carbon materials relies on structural descriptions that imply heterogeneity at the nanometric scale due to the tendency of carbon atoms to form sp^2 aromatic clusters. In this context hydrogen plays a key role in determining the microstructure and the optical properties. The presence of aliphatic carbon bonding in ACH2 is in agreement with the general result that hydrogen favors the sp^3 carbon hybridization and reduces the degree of aromatic sp^2 clusters in hydrogenated carbons (Robertson 1991).

The decrease of the 3.4 μ m band reported in the previous section is evidence for a reduction of the hydrogen content in ACH2 upon irradiation. Among the different mechanisms that can produce band reduction, photodissociation of CH bonds plays a primary role since the photon energy used in our experiments is larger than the dissociation energy for CH bonds (e.g. Iida et al. 1984). In Paper I several possible mechanisms for the reduction of the C–H stretching mode (in that case, of simple species) of hydrocarbon material covered with ice were discussed. We follow the conclusion in Paper I that dehydrogenation should be responsible, since we do not see any evidence in our spectra for the introduction of oxygen containing groups, neither is there any indication from our three experiments that the ice layer influences the efficiency of UV processing (Table 1).

Hydrogen loss from hydrogenated carbons induces significant rearrangements of the carbon bonding configuration. To get insight in this transformation, it is worth comparing the spectral IR modifications induced in hydrogenated carbon grains by UV photons with those produced by heat treatment (Mennella et al. 1996b). In the latter case the intensity of the stretching band corresponding to sp³ CH vibrations in CH_{2,3} groups becomes less and less pronounced with temperature, while the aromatic CH stretching mode at 3050 cm⁻¹ increases in intensity as the temperature increases: at 515 °C the aromatic mode dominates the spectrum and only a weak band at 2925 cm⁻¹ is present. Unlike heat processing, the transformation of CH bonds into aromatic CH bonds is not observed during UV processing.

The different behaviour of the CH stretching modes is consistent with the structural variations deduced from UV-Vis spectroscopy, optical gap variations, and Raman spectroscopy (Mennella et al. 1995, 1998) and provides a self consistent scenario for the evolution of hydrogenated carbon grains. Both the processes induce an increase of the sp² aromatic clustering degree. In the case of thermal annealing, the increase in number is accompanied with a growth of the graphitic clusters, especially after the complete hydrogen effusion, at temperatures higher than 600 °C. On the other hand, UV photons give rise to a lower dispersion of the graphitic cluster size; see Mennella et al. (1998) for further details.

4. Astrophysical implications

To analyze the implications of the present work for the nature of the 3.4 μ m band carrier, we have to estimate the UV processing degree for interstellar hydrogenated carbon grains. Using as a reference the interstellar radiation flux, 8 10⁷ photons cm⁻²s⁻¹ (Mathis et al. 1983), the laboratory fluence of 2.8 10¹⁹ photons cm⁻², leading to a reduction of the 3.4 μ m band intensity of 94%, corresponds to an exposure time of 1 10⁴ yr. This time is much shorter than the typical time-scale of 3 10⁷ yr that grains spend in the diffuse cloud medium before entering dense regions. If our photodestruction of CH bonds is representative of processing occurring in space, then the CH aliphatic component should be absent in the spectra of diffuse lines of sight, in contrast with observations.

The presence of the aliphatic 3.4 μ m feature implies that a mechanism able to reform CH bonds must be active in diffuse regions. This mechanism has been identified with the interaction of carbonaceous particles with atomic hydrogen that is abundant in diffuse clouds. In Paper I, it has been estimated that the hydrogenation efficiency, f, (the ratio between the number of CH bonds formed and the number of H atoms impinging on the grains) necessary to balance the CH bond photodissociation by UV photons is ~3%. The efficiency f was computed using a photodestruction cross section $\sigma_{des} = 5 \ 10^{-19} \ cm^{-2}$ per photon. Following the approach of Paper I, the lower value of σ_{des} we have found for the hydrogenated carbon grains studied in the present work gives rise to a value for f less than 1%.

Mennella et al. (1999) have recently studied the interaction of carbon grains with atomic hydrogen. They have found that the 3.4 μ m aliphatic mode is activated in nano-sized hydrogen-free carbon particles by exposure to a beam of H atoms. The laboratory H fluence was a factor 0.01 of that expected in a diffuse cloud time scale. The feature fits the interstellar band very well and the estimated hydrogenation efficiency is f = 0.06. This value is larger than the value required to balance the photodestruction. Therefore, we conclude that under diffuse medium conditions the formation process prevails. This conclusion is compatible with the presence of the aliphatic band towards diffuse medium lines of sight.

Our laboratory results indicate that dehydrogenation of the carbon particles by UV radiation, with a consequent reduction of the 3.4 μ m band intensity, can still proceed despite an ice layer. On the contrary, the presence of the ice mantles prevents the hydrogenation by H atoms of the underlying carbon particle (see, for a detailed discussion, Paper I). Thus, the destruction of CH bonds by UV photons prevails over the formation by H atoms under dense medium conditions. We have to verify whether in these conditions UV processing of hydrogenated carbon grains can be responsible for the absence of the aliphatic CH stretching mode. From the observational point of view a band intensity $\tau(3.4 \ \mu m) < 1 \ 10^{-2}$ would be difficult to detect in the spectra of embedded protostars (for example see Fig. 2 in Brooke et al. 1996). The intensity of the feature must be reduced at least to this detection threshold going from diffuse to dense medium. Of course the reduction depends on the initial intensity of the band. As a reference we assume the intensity, $\tau(3.4 \ \mu m) = 0.22$, observed in the diffuse medium toward the Galactic Center source IRS 6E. Under the limit assumption that all of the carriers of this feature are incorporated in a dense cloud, a destruction of 95% would account for the non detection of the feature in dense regions. The CH bond photodestruction cross section for hydrogenated carbon grains estimated in the present experiments indicates that the UV fluence necessary for such a destruction is $F_{\rm d} \sim 3 \ 10^{19}$ photons $\rm cm^{-2}$.

Inside a cloud the internal UV flux of $\sim 1 \ 10^3$ photons $\mathrm{cm}^{-2} \mathrm{s}^{-1}$ prevails over the shielded external radiation field (Prasad & Tarafdar 1983). The internal flux can only contribute to a 10% reduction of the band during the cloud lifetime. Therefore, the driving factor for grain evolution in dense regions is the attenuation of the interstellar radiation field necessary to allow the formation of an ice mantle and the destruction of the CH bonds. Under quiescent conditions, H₂O ice accretion starts once the line of sight visual extinction exceeds $\sim 3 \text{ mag}$ (Whittet 1996). This extinction threshold refers to the entire cloud. Thus the attenuation of the interstellar radiation field for a grain inside the cloud is at most equal to half of the extinction threshold. Under these conditions the UV flux is reduced to $F \simeq 5 \ 10^6$ photons cm⁻² s⁻¹. To obtain the fluence $F_{\rm d}$ grains should reside at the edge of the cloud for $\sim 2 \ 10^5$ yr which corresponds to about 1% of the cloud lifetime of $\sim 3 \ 10^7$ yr. As discussed in Paper I, circulation of material in dense clouds and the enhancement of the penetration of UV photons in clumpy and filamentary structures may cause the intense short term exposures able to account for the dehydrogenation of carbon particles.

On the other hand, cosmic rays may penetrate deep inside a dense cloud and interact with carbon particles, providing a possible further contribution to the 3.4 μ m band evolution. Several experiments have been performed with the aim to study the chemical modifications induced by ion processing on carbon based materials (e.g. Strazzulla 1998; Moore 1999). However, to the best of our knowledge, no specific measurement has addressed the problem of the 3.4 μ m band modification on samples similar to those analyzed in the present work. This matter will be the subject of a future laboratory study.

5. Conclusions

The present experiments show that the 3.4 μ m band is almost completely reduced in hydrogenated carbon grains upon UV irradiation under simulated diffuse and dense conditions. UV photolysis is the dominant factor for the destruction of CH bonds. The derived photodestruction efficiency indicates that interstellar hydrogenated carbon grains will be dehydrogenated by UV photons in the interstellar medium. In the diffuse medium this process is counteracted by hydrogenation of H atoms interacting with carbon particles. Actually, hydrogenation must exceed dehydrogenation, since we observe the band.

On the other hand, inside dense clouds grains will be covered by ice mantles and hydrogenation of the underlying carbon particle will be inhibited (Paper I). It is expected that a gradual photodehydrogenation of grains takes place. Our results indicate that the absence of the 3.4 μ m feature in dense regions may be accounted for if grains with an ice mantle are processed by ~3 10¹⁹ photons cm⁻² during the cloud lifetime.

The decrease of the aliphatic CH stretching mode is achieved in our experiments without destroying the carbon particles. Therefore, the proposed scenario is compatible with the transformation of interstellar carbon particles, whose aliphatic component is destroyed and (re)formed in dense and diffuse medium, respectively.

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