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SIZE EFFECT IN OPTICAL SPECTRA OF AMORPHOUS CARBON GRAINS(*)

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1. Introduction

The interstellar extinction curve presents a trend which can be explained only by a limited number of materials, such as carbon, graphite, SiC and so on[1-4].

Amorphous carbon (AC) is becoming an important candidate for explaining interstellar dust composition[5,6,7]. In fact, extinction spectra of AC grains are characterized by a broad band around 240 nm[7,8], similar to that observed in the interstellar extinction curve.

In addition the spectrum shows in the near infrared many bands which match those commonly named as the "unidentified interstellar bands"[8].

We report extinction spectra of AC grains measured in the spectral range 100-800 nm. These measurements show a main bump at about 235 nm and a few features at shorter wavelengths, which can be explained by a size effect on the dielectric constant of the material.

2. Experimental

The production method of AC grains is extensively described elsewhere[8]. The particle were collected on LiF substrate (cutoff at 115 nm) to allow UV transmission measurements. Transmission electron microscopy on the same particles shows chain-like structures (Fig. 1) composed by grains having spheroidal shape and average sizes of about 100 Å.

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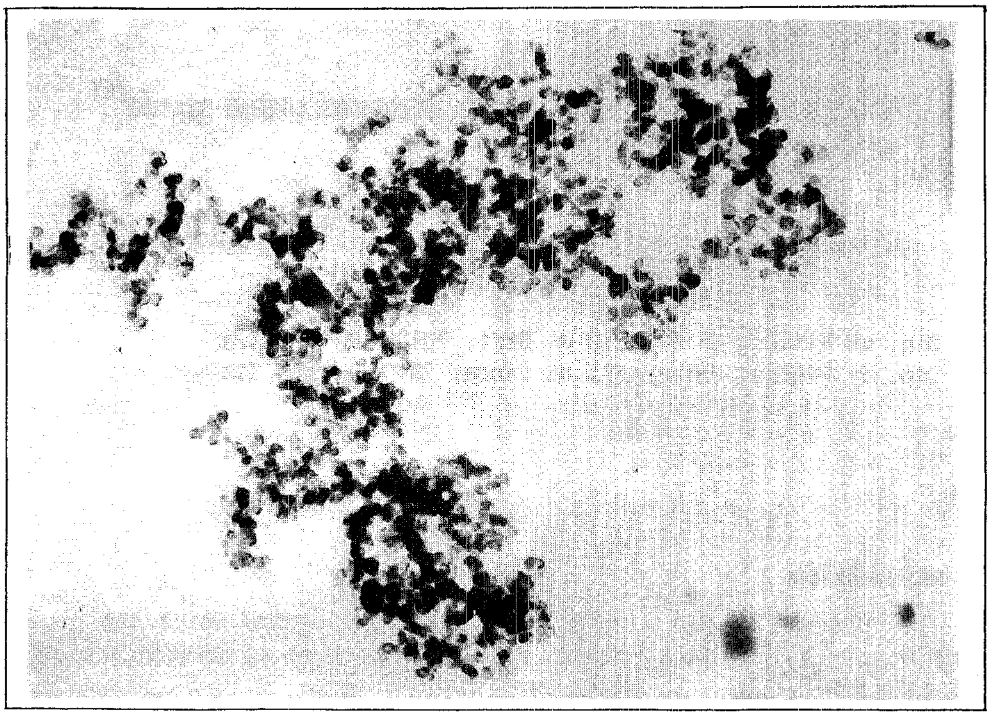

Fig. 1 - Electron microscope photograph of an amorphous carbon grain sample. The particles appear to have spheroidal shape.

The ultraviolet (UV) extinction measurements, from 100 to 300 nm, were performed, at room temperature, at the PULS synchrotron light facility of the Frascati National Laboratories (INFN).

The long wavelength region (200-800 nm) measurements were carried out by using a double-beam spectrometer. The two spectral range overlap to allow matching of experimental data.

3. Results and discussion

Fig. 2 shows the extinction normalized to the mean particle radius $\bar{a}$, $Q_{ext}/\bar{a}$ for the grains collected at 10 cm from the arc. The main features of the spectra are a wide band centered at 235 nm, a shoulder at about 180 nm and a well pronounced peak at 150 nm followed by a steep increase of extinction at shorter wavelengths. At wavelengths longer than 300 nm the extinction decreases following a $\lambda^{-\alpha}$ power law with α=1.4 for all the samples. This behaviour is expected for an amorphous structure: in fact, it is well known that amorphous materials show a decrease in extinction below the absorption edge, less steep than the corresponding ordered material. Graphite which is one of the ordered structures of carbon, is characterized by $\alpha=2$[9].

The above mentioned structures depend on the collection distance from the arc, beeing more pronounced at larger distance (see Fig. 3). The smearing of the peaks at the shortest distances may be due to the fact that the samples contain more particles per unit area, and therefore, the single grain properties become less evident. Furthermore, the peak position shifts towards shorter wavelengths for samples collected at a shorter distance from the arc.

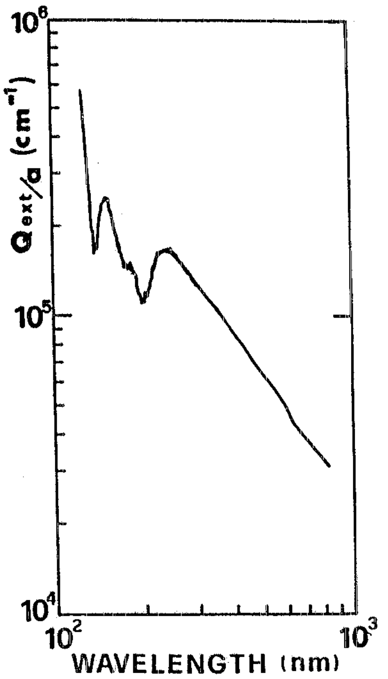

Fig. 2 - Extinction spectrum at 300 °K of amorphous carbon grains collected at 10cm from the arc discharge. The two overlapping spectra at low and high energy are measured by a visible spectrometer and by the UV synchrotron light radiation respectively.

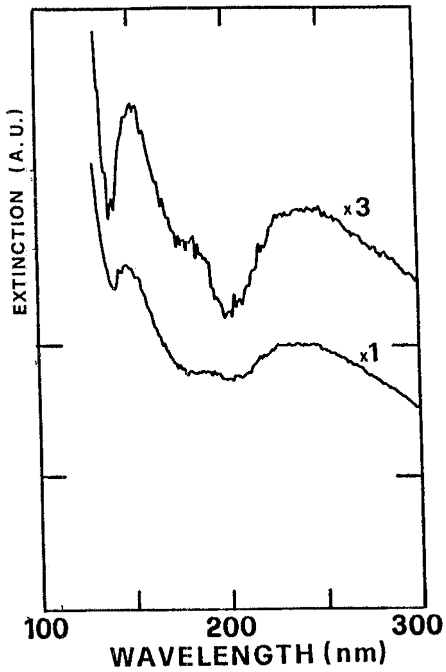

Fig. 3 - Ultraviolet absorption spectra measured for two different amorphous carbon grains sample, collected at 7 cm (a) and 10 cm (b) from the arc discharge. The spectral resolution is 8 Å. Relative intensity factors of each curve are indicated.

The band at 235 nm (5.28 eV) in AC samples is attributed to a surface plasmon resonance[10] and it corresponds to the similar band, located at 220 nm, of graphite. This identification is furtherly supported by the absence of any absorption structure at energies lower than 5.28 eV. The different position between the two solid state structures could be due to the different interatomic distances of atoms in graphite and AC grains.

The structures observed above the surface plasmon band are novel for carbon grains. They can be ascribed to quantum size effects, which become important when the grain is constituted by a small number of atoms. In fact, assuming a density of 2 g/cm^3 and a mean interatomic distance of 2.6 Å, the number of atoms per grain is of the order of 10^4.

Several theoretical models concerning the size effect have been published[11-13]. The results of the calculation give dielectric constant function which is more and more structured when the grain size decreases. Moreover, the position and the widths of the structures depend on the grain size and also on the embedding medium[13]. Particularly, by decreasing the grain size, the peaks can shift both to lower or higher energies depending on the surface potential[13].

In our experimental case, we have no detailed direct information on the dependence of the grain size on collecting distance. In spite of this fact, it is known that for AC particles produced by arc discharge collisional growth is active, and therefore, particles collected at longer distances are expected to have larger sizes. Consequently, we can explain the observed shift of the peaks as an effect due to size change of particles. In the particular case, we have that the peaks shift to higher energy by decreasing the grain size.

Theoretical calculations give this dependence and the opposite one too, depending on the embedding medium. As reported in ref.(13), the influence of size on the energy shift of the peak depends critically on the parameter Z, which is a measure of the surface potential. When the embedding medium is vacuum, Z is positive and according to Figs.10 and 12 in ref.(13), the observed blue shift is an indication that interband transitions give a noticeable contribution to the absorption spectra. This is in agreement with the fact that the steep rise of the absorption below 140 nm is due to electronic transitions to excited states. In this view, the observed structure can be considered as a modulation of the low energy absorption tail due to the size effect.

The other size effect which has been observed and calculated is a broadening of the features[11,13]. In our spectra there is some indication of this characteristics, but a quantitative analysis is not possible. A more detailed investigation is necessary to assess this last point.

From the point of view of the interpretation of the observed interstellar extinction, we note that published data do not seem to indicate the presence of extra features beside the usual one at about 220 nm. From our results it can be derived: 1) if amorphous carbon may be considered a likely cosmic dust candidate material, the particles should be very small, i.e. $\langle a \rangle < 50$ Å, in order to show the bump at wavelengths shorter than 235 nm; 2) the absence of the other peaks suggests that, in the interstellar space, the particles should be gathered in dense patches, rather than uniformly dispersed along the line of sight.

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