FREE ELECTRON LASER STUDY OF FREE CARBON CLUSTERS

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Abstract

UV absorption from carbon nanoparticles is a very interesting astrophysical topic. The prominent hump centred at 217.5 nm is the most dominant feature in the interstellar extinction curve and also the most controversial and a long-standing problem in astrophysics. At the University of Milano an experimental set-up based on a Pulsed Microplasma Cluster Source has been developed for the investigation of free clusters at the Elettra Gas Phase beamline. The cluster source produces very intense cluster beams with tunable size distribution. The design of the apparatus is extended with a chamber for gas phase reaction (water vapour, CO, H_2...) providing a unique opportunity to study the gas phase properties of carbonaceous particles in different environments.

We plan to investigate Resonant Raman scattering of free carbon particles tuning the high brilliance UV/VIS storage ring FEL of ELETTRA across the region of 217.5 nm where the UV absorption hump in astrophysical data is observed and where a number of electronic transitions exist for variable size linear carbon chains.

INTRODUCTION

Since its discovery by Stecher (1965), the ultraviolet extinction curve feature observed at 217.5 nm has been attributed to π-electron plasmon absorption or π-π * band transitions in small graphite particles or amorphous carbon grains. Despite the early assignment to carbonaceous material, the exact physical nature of the carrier is still unknown and strongly debated. One peculiar behaviour of the 217.5 hump is the constancy of its spectral position regardless of the choice of the astronomical object under observation [1]. On the other hand, the peak position of the bump predicted for graphite particles is quite sensitive to grain size, shape and adsorbate coatings, which is inconsistent with the observations.

Many different models have been proposed to explain the experimental observation and many different attempts have been made to reproduce in laboratory an adequate prototype for cosmic dust [2,3,4]. Experiments conducted on hydrogenated carbon nanoparticles isolated in noble gas matrix pointed out the relevance not only of particle shape and size but also of their chemical environment [1]. Other observations indicate that the presence of an ice mantel surrounding the carbon particles strongly influence their UV optical response [3]. Other authors have succeeded in synthesizing carbon aggregates that show optical constants fitting quite well the extinction curve [4, 5], but, the adopted methods don't match very well with the environment conditions where carbon grains in the intergalactic medium are expected to form.

Actual models of dust astrophysics lack of experimental data about carbon dust in gas phase, their optical proprieties, mass distribution, reactivity and of course their absorption of the UV light in the 217 nm region. In order to attempt a meaningful reproduction, at the laboratory scale, of this astrophysical system it is important to study carbon clusters in isolated condition, as a function of their dimension and chemical environment. Recently the potential of optical spectroscopy in free jet expansion for experiments on astrophysically relevant species has been demonstrated [6].

Resonant Raman Scattering (RRS) is a powerful technique for the characterization of such a system. It combines the sensitivity to vibrational properties of carbon structures relevant for astrophysics like carbyne [7, 8] with a selective transfer of energy when the exciting photons are tuned at the energy corresponding to a given resonance, increasing in this way the scattering cross-section.

We present here an experimental setup for a RRS study of free carbon clusters which make use of the storage ring Free Electron Laser (SRFEL) radiation.

EXPERIMENTAL SETUP

This section quickly reviews the experimental setup that we are developing at ELETTRA in the framework of the collaboration between the CIMAINA of Milan University and the SRFEL group of ELETTRA, Trieste.

CESyRA

CESyRA (Cluster Experiments with Synchrotron RAdiation) [9] is a research project of the CIMAINA focused on free cluster spectroscopy with the high intensity UV and soft X-ray light from the ELETTRA synchrotron radiation facility.

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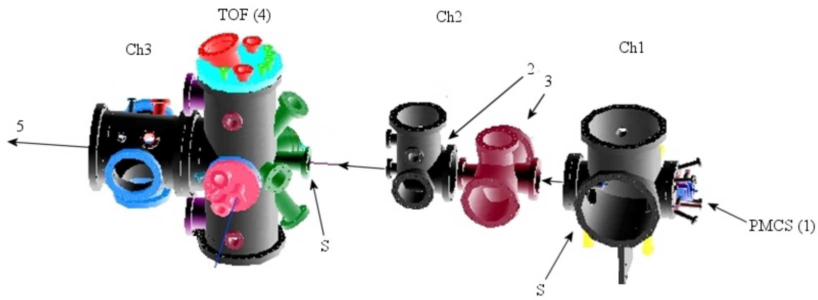

Fig. 1: Main elements of CESyRA apparatus: (Ch1) expansion chamber, (Ch2) deposition and gas exposure chamber, (Ch3) TOF, (1) PMCS, (2) manipulator, (3) gas line, (4) TOF, (5) to dumping chamber and quartz microbalance.

The experiments are implemented in a UHV compatible supersonic cluster beams apparatus. The main constituents of the system are sketched in figure 1. Briefly, it consists of three differentially pumped, high-vacuum chambers (Ch1, Ch2 and Ch3) separated by two skimmers (S). Heart of CESyRA apparatus is the Pulsed Microplasma Cluster Source (PMCS) [10,11], developed at the Molecular Beam and Nanocrystalline Materials Laboratory in Milano, which is able to deliver highly collimated and intense seeded beams of clusters from refractory materials (typical deposition rate for carbon: 100 µm/h at 500 mm source-substrate distance, ~ 0.8 cm^2 covered area). The PMCS is based on target ablation obtained by He plasma sputtering: a pulse of He flux is directed against a target by means of a valve driven by an electromagnet (opening time of about 300 µs). The gas is then ionized by a pulsed discharge fired between the target rod (cathode) and the anode. The sputtered particles are carried through an aerodynamic lens system by the He flow and the mixture eventually undergoes a supersonic expansion into a high vacuum chamber. In order to prevent the formation of holes on the cathode rod and to reduce the need for source maintenance, since the ablation is extremely localized, the target rod is maintained in rotation during source operation (see figure 2). The clusters are emitted in pulses that propagate at a speed of about 1000 m/s and that last typically 15 ms. Lighter aggregates are mostly located at the head of the pulse while the heavier ones at the tail.

The PMCS is located outside vacuum and communicates through the nozzle with the expansion chamber (Ch1). The beam passes through a skimmer that, due to particle focusing on the beam axis [12], efficiently separates the clusters from the carrier gas. In the second chamber a gas cell is installed to allow gas exposure of the free particles in the beam (3). A diaphragm separates the second from the third chamber (2) to produce differential vacuum and finally the beam is directed into an interaction chamber where a short linear time of flight (TOF) mass spectrometer is mounted perpendicularly to the plane formed by the intersection of the light and cluster beams (4). A sample manipulator is mounted, in order to intercept the beam and thus to allow cluster deposition onto a substrate. Finally, the beam is dumped onto a quartz microbalance (5) that monitors the cluster flux.

The SRFEL of ELETTRA

The CESyRA setup has primarily been thought for application with synchrotron radiation on refractory materials in gas phase, demonstrating for the first time the possibility of performing XAS measurements on free Ti nanoparticles [13]. However, for this particular application on carbon clusters, the requirements on the light source that has to be used make the SRFEL of Elettra [14] the best candidate for our objective. To probe the cluster beam in gas phase, we need a very high flux of photon with a wavelength that can be precisely tuned around 217.5 nm (5.7 eV). In this respect, the Elettra SRFEL can produce a very bright monochromatic beam that can be directed into the interaction volume by means of few optical elements, minimizing the photon flux losses due to radiation transport.

The maximum average lasing power that can be achieved using a SRFEL is limited by the heating of the electron beam induced by the laser onset [15, 16]. The increase of the electron-beam energy spread is indeed responsible for the diminution of the optical gain while, at

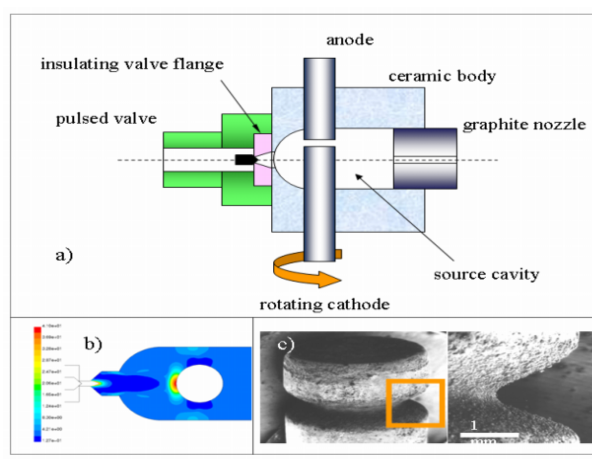

Fig. 2: a) schematic diagram of the PMCS source; b) pressure contour plot of He jet inside the PMCS cavity (see ref [11]); c) scanning electron microscope micrographs of the cathode region eroded by the plasma. The cathode is continuously rotated in order to prevent the formation of holes. A very smooth and precise trace formed by plasma erosion is clearly distinguishable.

saturation, the latter reaches the level of the optical cavity losses. However, for applications requiring a high peak power, the FEL power can be ‘‘concentrated’’ into a series of giant pulses, applying the so-called Q-switching regime. In this case, the peak power is considerably enhanced (one to few orders of magnitude) while the average power is only slightly reduced [17]. To operate the SRFEL in Q-switching, we apply a modulation on the radiofrequency (RF) that drives the electron bunches in the storage ring (figure 3a) [18]. The giant pulses that we obtain are typically 100 μs long (figure 3c). The development of a giant pulse introduce a quite big energy spread that implies a recovery time for the dumping of the electron beam of the order of 10~100 ms. For this reason Q-switching is normally operated at a repetition rate of few Hz (figure 3a and 3b). In RRS experiment setup, the RF modulation is triggered after a variable delay by the opening of the He valve in the clusters source. Adjusting the delay we are able to probe, with the highest power available, different parts of the cluster pulse and therefore sample cluster populations with different average size.

In order to probe the clusters with the Raman scattering in the spectral region of interest, we use mirrors for the FEL optical cavity especially designed for that wavelength. The interferential coating has been prepared at the Laser Zentrum Hannover.

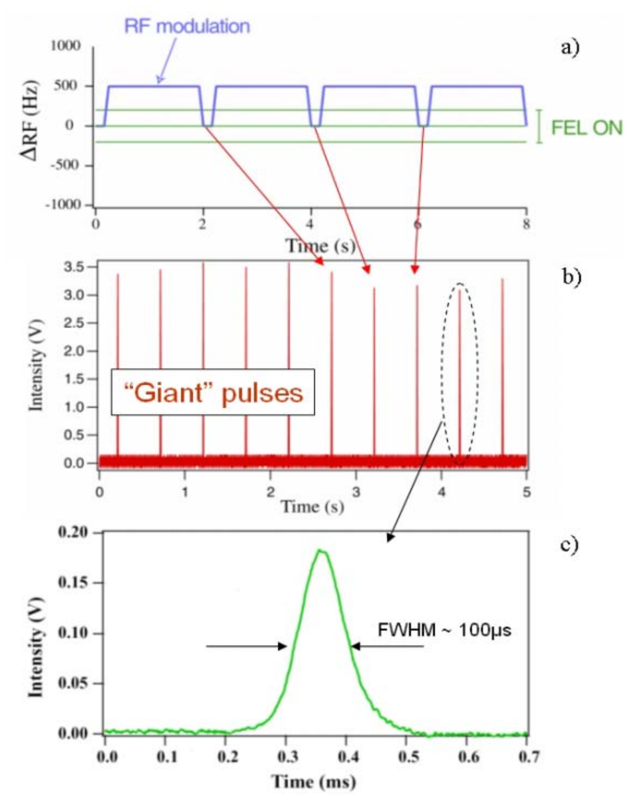

Fig. 3. Q-switching of the SRFEL: a) the RF modulation can be triggered by the opening of the He valve of the clusters source, b) development of a sequence of giant pulses, c) detail of a giant pulse: typical duration is 100 μs.

PRELIMINARY TESTS AND FUTURE DEVELOPMENT

The CESyRA system is in the experimental hall of ELETTRA since May 2006 for the long term project approved on the Gas Phase beamline [9,13,19]. When the chamber is not in use for this project it is available for the preparation of the RRS experiment.

In Raman spectroscopy, one of the most frequently encountered problems is related to the presence of the elastic background. Since the Raman cross section is typically 4-5 orders of magnitude smaller then the elastic one (Thomson scattering), the Raman signal risks to be submerged by the tails of the elastic peak. Moreover, in our setup, the interaction volume is in vacuum, separated from the spectrometer and the SRFEL by quartz view-ports. This introduces an additional source of scattered light that further decreases the "peak to background" ratio. The first step in the preparation of the Raman experiment has been the measurement of a total scattering yield, i.e. the collected light signal regardless of its spectral distribution, in order to evaluate the relevance of the background and to assess the interaction between the clusters and the radiation. For this purpose, we used a photomultiplier tube (PMT) to detect the photons that are collected by a small quartz lens focusing in the interaction volume about 5 mm far from the PMCS nozzle, where the cluster density is close to the maximum. When the setup will be optimized, the PMT will be replaced with a spectrometer.

Since now, few hours have been dedicated to optimize the experimental setup. FEL light at 217.5 nm has been generated with a very good stability. We expect to extract a power of few tenth of mW in free-run mode that should correspond to a flux of 10^{17}~10^{18} ph/s in the Q-switching regime. The FEL light has been used to perform the alignment of the chamber as well as some preliminary tests aimed at optimizing the Raman setup. The light was focused on the cluster beam by means of a lens located outside the chamber and one mirror in vacuum at 45º that deflect the light in the vertical direction. A second mirror at 45º was used to extract the light from the chamber. The focus of the collecting lens was placed at the intersection of the laser and clusters beams, so that the collection was done in the direction perpendicular to both of them. A multichannel time to digital conversion board has been used to record the detected events keeping track of their delay with respect to the opening of the valve. The full range temporal window of detection is 20 ms, with a time resolution of 82 ps. The acquisition software allows integration over an undefined number of consecutive pulses. To obtain a good background subtraction despite the possible drifts in laser and cluster source operation, the measurements have been done alternating acquisition of pulses with and without clusters; this was obtained by firing the vaporization discharge in the PMCS every second gas pulse only. Typical integration time was 5 min at a repetition rate of 5 Hz that corresponds to acquisition

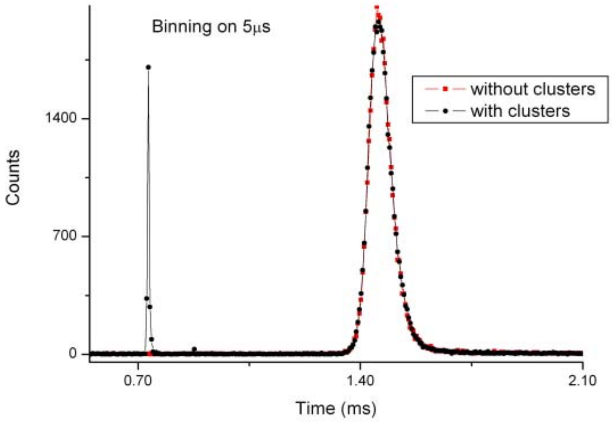

Fig. 4: Acquisition with PMT of the scattered intensity. Black line (circles) SRFEL pulse plus clusters pulse. Red line (square) SRFEL pulse with only the carrier gas (notice the absence of the HV discharge noise).

of about 750 laser pulses with clusters in the beam and 750 without clusters.

Figure 4 shows an example of detected events for an acquisition obtained setting a nominal delay of 1.2 ms between the He valve and the RF modulation. The peak at 1.45 ms corresponds to the photon counts collected under the giant pulses of the SRFEL. The black line with circles is associated to the pulses with clusters and the red one (squares) without clusters. The sharp spike at 0.73 ms is due to electrical noise introduced by the HV discharge in the PMCS and thus occurs only in the pulses where the generation of clusters is on; excluding this instrumental effect, no significant differences were measurable between the two curves at any delay. An expanded view of the peak at 1.45 ms (figure 5) shows that photon counts are detected with a time structure that closely follows the microtemporal structure of the FEL radiation. The fact that the detection rate is of the order of one photon count per micropulse indicates that the intensity of light scattered inside the chamber by the optical elements is hindering the signal of interest as the detector is probably saturated by background counts.

Those preliminary results show the importance of screening the detector from the intensity scattered by the optical elements. At present, we are developing a new optical scheme to minimize this source of background that certainly would prevent the detection of the weak Raman signal. We are also implementing a Nd:YAG table top laser that, even if not tunable at the proper wavelength, will be helpful for off-line preparation of the experiment with the SRFEL.

Considering the small number of shifts that for the moment have been dedicated to the experiment, we can be optimistic for the future developments. The CESyRA apparatus and the SRFEL reveal to be stable enough for the acquisition of data with good statistic.

We acknowledge the storage ring control group of Elettra for collaboration and Stefan Günster of LZH (Hanover) for the realization of FEL optics.

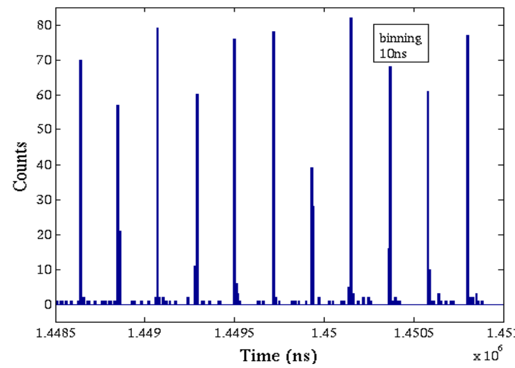

Figure 5: Expanded view of the microtemporal structure of photon count peak under FEL giant pulse in the Q-switch mode.

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