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Spectroscopic investigation of refractory molecules produced by laser vaporization

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ABSTRACT

In classical ovens, vapour pressure of small molecules including refractory compounds is generally too weak to undertake their spectroscopic characterization in the gas phase. Laser sources as they allow any material to be vaporized whatever its melting point, bring a satisfactory answer to this problem.

We designed a source of this type with the aim to undertake the spectroscopic characterization of small molecules including carbon or nitrogen together with a metal of large astrophysical abundance (Al, Mg, Na, Fe).

The first mass spectra of neutral molecules as well as ions produced by this source are presented in this paper.

INTRODUCTION

The observation of any molecule in astrophysical media relies on their preliminary characterization in the laboratory. For this reason, spectroscopic analysis of more and more complicated molecules, in connection with astronomers' observations, continues to be very fruitful for molecules containing the more abundant elements such as H, C, O, N and S. The situation is less favourable for metal-bearing molecules where observations in astrophysics as well as in the laboratory are less developed. They are mainly limited to molecules where metal atoms are associated with O, H, S and, except some noticeable examples like MgCN¹, very less is known about molecules including metal and N or C atoms. Obviously, this situation is due to the refractory character of those molecules that makes difficult the yield of large amounts in the gas phase.

In his wide review on the observations and chemistry of refractory elements in the interstellar medium, Turner² came to the conclusion that a large laboratory effort is awaited to characterize carbides but especially nitrides of Al, Na or Mg that could appear preferentially than the corresponding oxides in some star-forming regions of the interstellar medium. Moreover, after the observation of SiC in IRC +10 216³, mono or polycarbides of Al, Mg or Fe are expected to be observed too.

We present in this paper a laser vaporization source, especially designed to produce small molecules of refractory materials. Our preliminary results demonstrate the capability of this source to produce neutral molecules (AlC and Al_nC), as well as ions directly ejected from the laser vaporization plasma (Fe_n⁺ and C_n⁺). We shall give an idea of the investigations that will be conducted in

forthcoming experiments with this source.

EXPERIMENT

Fig. 1. The vaporization source.

Fig. 2. Two photon ionization.

In a pioneering work by Smalley, the efficiency of laser vaporization sources to produce large amounts of refractory compounds in the gas phase has been demonstrated for any metal, up to the most refractory like molybdenum and tungsten ⁴.

Briefly, a target rod is located in the throat of a pulsed nozzle (Fig.1). It is shot by the second harmonic of a pulsed YAG laser leading to metal vaporization and formation of a high temperature plasma. Before laser pulse, helium was injected into this source through a pulsed valve in such a way that the laser shot coincide with the maximum pressure of the helium carrier gas; the injection pressure can be varied in the 1-10 bar range. In the waiting room (0.2 cm³) begins the expansion of the laser plasma into the carrier gas with recombination and thermalization of the plasma and production of small molecules and clusters.

After the nozzle, these molecules and clusters continue to expand into the vacuum (10⁻³ torr), forming a cool molecular beam. A skimmer placed 1-2 cm downstream allows this supersonic molecular beam to enter the low pressure region (10⁻⁶ torr) where various excitation processes will be undertaken with different lasers. In any case, laser excitation results in the yield of ions. These ions are produced into the extraction region of a time of flight mass spectrometer (TOF), that permits mass characterization.

The simplest excitation is the direct ionization by one UV photon. Increasing the energy of this photon, new peaks of given masses appear as the photon energy

reaches the ionization potential. By this way, ionization potentials can be easily determined. Moreover, photoionization efficiency as well as its evolution near the threshold can also be studied. In another scheme depicted in Fig.2, one photon excites the molecule to an electronically excited state while a second photon brings it beyond the ionization limit. By scanning the exciting photon over a given electronic state an ion signal is produced only when the exciting energy matches a v, J level of this state, allowing its spectroscopic investigation.

PRELIMINARY RESULTS

We started the experimental program, with an aluminum rod, by the mass characterization of molecular species existing in the molecular beam. This was achieved by direct ionization with one UV photon provided by a frequency doubled excimer pumped dye laser. A typical spectrum is shown in Fig 3 with a photon energy: 5.6 eV. This spectrum exhibits a large series of Al_n clusters, up to $n=40$, as they are usually produced in this kind of source ⁵.

Fig. 3. Mass spectrum obtained by laser vaporization of an aluminum rod combined with direct one photon ionization.

As discussed in a former paper ⁶, the most exciting feature of this spectrum is the appearance of mass peaks for the Al_nC molecules. The large intensity of the AlC mass peak gives us the opportunity to start with spectroscopic investigation of this molecule using the one photon resonant-two photon ionization

technique and scanning the resonant photon over AlC electronically excited states. Moreover, we intend to undertake a systematic study of the ionization potential and photoionization efficiency of Al_nC molecules. We expect valuable information regarding the C-bonding with metal particles to be deduced from these experiments.

Fig. 4. Mass spectrum obtained by laser vaporization of a graphite rod.

In another way, by replacing the ionization laser by a high voltage pulse, we show up (Fig 4), by making use of a graphite rod, that large amounts of carbon ions, produced in the laser vaporization plasma, live all along the molecular beam. As a consequence, their spectroscopic characterization, by laser induced fluorescence, will be possible just downstream the nozzle.

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