

# Developments and Plasma Studies at the ATOMKI-ECRIS

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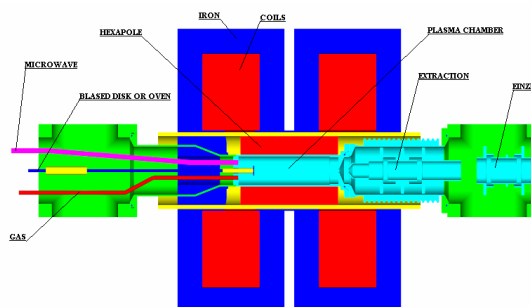
**Abstract.** The 14.5 GHz ECR ion source of the ATOMKI is a stand-alone device producing highly charged ion beams for ion-surface experiments and a variety of low charged plasmas and beams for plasma physics studies and for practical applications. In the past two years we performed plasma diagnostics measurements using Langmuir-probes and X-ray camera. Langmuir-probe results allowed estimating the plasma potential close to the resonance zone. The studying of X-ray pictures of Xe-Ar plasmas helps understanding the gas-mixing phenomena. A mixture plasma of fullerene and ferrocene was generated and FeC<sub>60</sub> hybrid molecules were detected in the extracted beam.

## INTRODUCTION

In the ATOMKI a 14.5 GHz electron cyclotron resonance ion source (ECR, ECRIS) operates as a stand-alone device to produce variously stripped plasmas and low-energy ion beams [1,2]. So far it delivered H, He, N, O, Ar, Kr, Xe (from gases) and C, C<sub>n</sub>, C<sub>60</sub>, F, Fe, Ni, Zn and Pb (from solids) plasmas and beams. During the last few years several improvements have been carried out on the ECRIS. The mirror ratio of the magnetic trap was increased by inserting additional soft iron plugs at the injection side. A movable, biased electrode was installed to tune the plasma potential and to effect on the extraction conditions. More recently this electrode was equipped with water cooling while it is still biased and movable on the high voltage platform. By this we expected to decrease the outgassing and so that we can apply higher microwave powers. The result of the first tests of the disc cooling is however, strange. It remarkably improved the intensities and charges for the case of xenon (we obtained Xe<sup>27+</sup>), but showed almost no effect on argon. The installation of a water cooling system for the puller electrode and placing soft iron plugs at the extraction side, are also under way. In Figure 1 the cross sectional view of the ion source is shown.

One of the specialties of our source it is equipped with three vacuum motion feedthroughs. One moves the first group of electrodes of the extraction system (puller and pre-lens). Another one is normally used

for the biased electrode which in our case has a disc shape or sometimes star (or triangle) shape. For oven experiences the biased electrode is changed by the oven so the oven in whole can also be moved and biased. The third feedthrough is used for Langmuir-probe diagnostics (not shown in the figure).



**FIGURE 1.** Layout of the ATOMKI ECR ion source. The internal diameter of the plasma chamber is 58 mm.

## PLASMA POTENTIAL

The measurement of space potential in laboratory plasmas is often necessary because information on electric fields, potential distribution etc. is frequently needed. The potential plays substantial role ECR ion

sources which combine magnetic mirror confinement of electrons with electrostatic confinement of the highly charged ions. Conventional Langmuir probes in ECR plasmas have been used to measure the plasma density [3,4]. To determine the plasma potential emissive Langmuir probes can be used. An emissive probe is essentially a biased hot wire probe, heated sufficiently to allow thermal emission of electrons. When the probe is biased negatively with respect to the local plasma potential, the emitted electrons can escape into the plasma and appear as an effective emission ion current. When the probe is biased to the local plasma potential, the emitted electrons are reflected back to the probe and the emission current disappears. There are several ways to calculate the plasma potential from emission probes characteristics. In our experiments we applied the simplest way. The U-shape emission probe was moved into the required position of the plasma chamber. First we developed the necessary heating power [5]. Then the probe potential was set to a value (we call as wall potential) where the emission current was suppressed to zero. This potential in most cases is very close to the plasma potential. Its changes caused by plasma modifications can be used to study the effect of external settings to the plasma. Further in this paper we simply call it as plasma potential.

Our first attempts at the ATOMKI-ECRIS and at the NIRS-KEI2-ECRIS [6] with classical U-shape emission probes (Current-Heated Emission Probes, CHEP) have been unsuccessful so far due to the limited lifetime of these types of probes. Nevertheless the advantage of these probes that they can be used in most places within the plasma chamber. We continue our efforts to find a better making and operation technique of CHEPs.

Then we decided to apply simple Langmuir probes heated to a high emission rate by the plasma itself. Obviously such probes can be used only in the hottest plasma region that is in the resonance zone (RZ). This is a spatial limit but the lifetime of these Plasma-Heated Emission Probes (PHEP) is long and if their size is small enough the plasma is almost undisturbed. We used 2.5 mm long and 0.4 mm thick cylindrical tungsten probe positioned to one of the estimated hottest plasma spots (as in Figure 3, later on). The holder of the probe was fit into thin ceramics tube.

We studied the variation of the plasma potential in the RZ with respect to external tuning factors as extraction voltage on/off, biased disc voltage and gas mixing. The ion source was tuned for  $\text{Ar}^{8+}$  or  $\text{Ar}^{11+}$  current. The magnetic field was less than the optimal to form a larger resonance zone and to position the

resonance zone exactly to the probe. Figure 2 summarizes the results. For additional information the  $\text{Ar}^{8+}$  ion current (in arbitrary units) is also shown in the figure. It belongs to the Extraction ON case.



FIGURE 2. The plasma potential at the resonance zone.

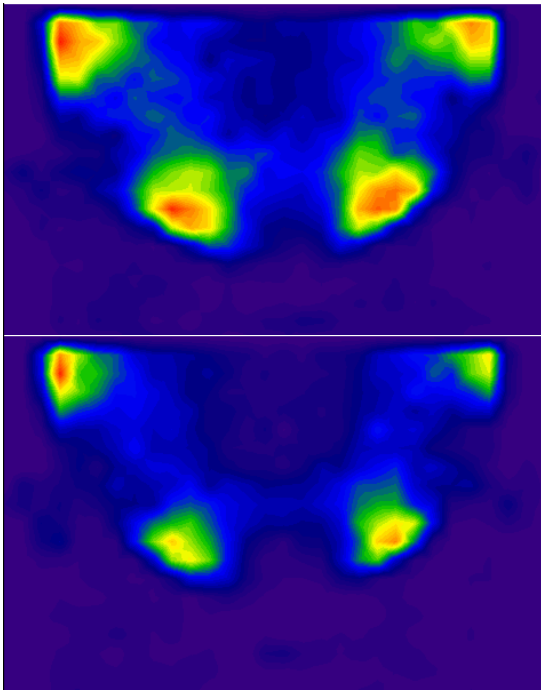
From the figure several conclusions can be drawn. The plasma potential decreases by both techniques (biased disc voltage and gas mixing) which increased the  $\text{Ar}^{9+}$  ( $q=8$  and higher) currents. Switching the extraction voltage off increased the plasma potential indicating a less-charged plasma. This means the charge state distribution (CSD) of the extracted beam does not mirror back completely the CSD in the plasma.

## X-RAY PICTURES

X-ray plasma images were made at the ATOMKI-ECRIS in 2003 using a pinhole and a high resolution CCD camera. This method had good spatial resolution as well as the capability of post-processed filtering of the images. During the measurements low and high charge state Ar, Xe and Fe plasmas were produced with simultaneous beam extraction. Full-size and selected region images were recorded and analyzed. The details of the measurement and some results of the prompt analysis were already published [7]. Since then no new measurement could be carried out however the analysis of the tremendous measured data was continued. Based on the CERN ROOT software package a method was developed to process and analyze the data. We concentrated to full-size images of xenon and xenon-argon mixture plasmas. In these

experiments the ECRIS was tuned for  $\text{Xe}^{3+}$  ions applying low microwave power (40 W). In the X-ray spectra strong xenon emission lines appeared between 4 keV and 5 keV originating from L shell transitions (from  $n=3$  to  $n=2$  bound state transitions) in different charge states of xenon. We developed a new post-processing technique to remove all of those parts from the X-ray pictures which can not originate from the xenon plasma as line emission. The details of this method were recently published [8].

Figure 3 shows a spatial distribution map of xenon in the plasma (upper image) and the case when argon gas is added to (bottom image). It is important to note that since the lifetimes of the transitions involved are usually short, line emission only occurs at places where energetic electrons to excite the ions are also present. The x-ray emission from xenon ions is the strongest where the overlap between the electron and the ion clouds is large. Our observation is that the high intensity xenon regions are more localized (more concentrated around high electron density regions) in the mixture case.



**FIGURE 3.** Comparison of spatial distribution of Xe ions in pure Xe plasma (up) and in Xe-Ar mixed plasma (bottom)

The indication for xenon ions being in higher charge states when mixing gas is added, is showed by

the charge state distribution of the extracted beam and by the shift of the xenon L peak position. In the mixture case there is strong shift of the line position as a function of the brightness of the spot. This appears only to a lesser extent in the pure xenon case.

The temperature of the ion cloud in the plasma is basically determined by the resulted depth of the potential dip of the electron and the ion clouds (generally around a few eV). The strong heating by the energetic electrons by coulomb collisions is balanced by the self cooling of the ion cloud due to the so called evaporation process controlled by the depth of the local ion trapping. Ions with energies higher than the potential dip leave the region removing energy from the local ion cloud providing the cooling effect. If additional cooling is provided by a lighter mixing gas ions can stay trapped longer and the cloud not only becomes colder but the ions will become further ionized.

The above observations and conclusions help a better understanding of the experimentally well-known fact that evaporative cooling of the ion cloud takes place in ECR sources. This results in a more localized and higher charged xenon ion cloud when the lighter argon gas is added. We must note however that these X-ray images were taken in case of low-charged plasmas. We consider important to repeat these type of measurements for the case of higher charged plasmas and we have plans to perform it in near future.

## IRON-FULLERENE PLASMA

In many laboratories new materials useful for nanotechnology and medical applications are searched and studied. In Atomki one of our future goals is to produce endohedral fullerene molecules (e.g.  $\text{N@C}_{60}$ ,  $\text{Fe@C}_{60}$ ) in large quantity. If this comes true, it will be possible to make building blocks for nano-parts, an ultra-contrast medium of MRI, and a magnetic nanoparticle for treatment of cancer.

For this experiment some modifications were carried out on the ECRIS similarly to [9]. The waveguide of the 14.5 GHz microwave generator was splitted and divided in order to couple very low powers (in the range of 1 watt or less) into the plasma. The iron plug at the injection side was removed. A low temperature oven was installed in the place of the biased disc. The oven can be biased and is on-line movable on the axes of the plasma chamber. The temperature was monitorized by thermocouple probe.

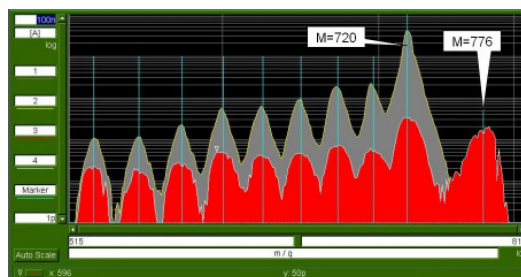
Usually 200...300 mg fullerene powder was placed into the stainless steel crucible of the oven. A simple MIVOC chamber was connected to one of the two gas feeding lines as close to the plasma chamber as possible. The chamber was filled with 200 mg ferrocene powder. The evaporation rate was controlled by careful external heating. The extraction voltage had to be kept as low as 600 V, because of the low mass-energy product of our bending magnet. So the extracted beam intensities were low.

First we developed independently the rough working conditions for single-charged dense iron and fullerene plasmas. Then a clean fullerene plasma was made. The temperature of the oven was about 450 C which corresponds to approx. 1 mbar local fullerene vapour pressure range. The bending magnet was set to the  $C_{60}$  peak ( $M=720$ ) and about 50...100 nA intensity of single-charged fullerene peak was obtained. Then the magnet was set to the position of the expected  $Fe@C_{60}$  or  $FeC_{60}$  peak ( $M=776$ ) and the ferrocene valve was opened. This usually caused strong instabilities in the plasma, all the fullerene peaks decreased and many new ones from the ferrocene appeared. A very difficult and long tuning was then followed. While the first ionization potential of the  $C_{60}$  and Fe are low (7.8 and 16.2 eV, respectively), obviously different working conditions are necessary to break the ferrocene and to ionize/excite the fullerene molecules. Finally we found reproducible setting to get a high peak around  $M=776$ . The key point of the tuning was the microwave power and the magnetic field strength at the extraction side. The typical power was several watts (sometimes several tens of watts) and the extraction magnetic peak had to be decreased down to the lowest possible value. At this value the resonance zone was very close to the extraction slit.

A typical  $C_{60}+Fe$  beam spectrum can be seen in Figure 4 together with an original (clean)  $C_{60}$  spectrum. The centre of the new big peak on the right side is at  $M=776$  which corresponds to  $FeC_{60}$  and/or  $Fe@C_{60}$  molecules. However the peak is wide and shows some structure. We think it may contain impurities attached to the  $C_{58}$ ,  $C_{59}$ ,  $C_{60}$  and  $FeC_{60}$  molecules. These impurities can be H, C and  $C_xH_x$  fragments ( $x=1...5$ ). Unfortunately the resolution of our bending magnet does not allow separating these components. Of course we can not tell at that moment the iron atoms are inside or outside the carbon cage, neither.

As a conclusion our experiment demonstrated that the ECR ion source generally can be used to produce mixed iron-fullerene plasma and  $FeC_{60}$  molecules

both in the plasma and in the beam. However, most probably other method (e.g. sputtering, electron bombardment) will be necessary to make a more clean iron component of the plasma.



**FIGURE 4.** Fullerene beam spectrum (upper curve) optimized for  $C_{60}^+$  ( $M=720$ ). Lower curve: fullerene+ferrocene spectrum optimized for the peak at  $M=776$ .

## ACKNOWLEDGEMENTS

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## REFERENCES

1. Biri S., Vámosi J., Valek A., Kormány Z., Takács E., Pálinskás J. Nucl. Instr. Methods B124 (1997) 427-430.
2. Biri S., Valek A., Kitagawa A., Muramatsu M., Proc. 15. Int. Ws. ECRIS. Univ. Jyväskylä, Finland, 2002 (JYFL Research Report 4/2002), pp. 49-52.
3. Kenéz L., Biri S., Karácsony J., Valek A., Nakagawa T., Stiebing K. E., Mironov V., Rev. Sci. Instrum. 73 (2002) 617-619.
4. Kenéz L., Biri S., Karácsony J., Valek., Nucl. Instr. Methods B187 (2002) 249-258.
5. Kenéz L. et al., paper under preparation.
6. Muramatsu M. et al, this proceedings.
7. Biri S., Takács E., Hudson L.T., Valek A., Radics B., Imrek J., Juhász B., Suta T., Szabó Cs., Pálinskás J., Rev. Sci. Instrum. 75 (2004) 1420-1422.
8. Takács E., Radics B., Szabó C.I., Biri S., Hudson L.T., Imrek J., Juhász B., Suta T., Valek A., Pálinskás J., 12th Int. Conf. on the Physics of the Highly Charged Ions (HCT'04), Vilnius, Lithuania, 6-10 September 2004.
9. Biri S., Valek A., Kenéz L., Jánossy A., Kitagawa A., Rev. Sci. Instrum. 73 (2002) 881-883.