LASER ABLATION OF SOLIDS INTO AN ELECTRON CYCLOTRON RESONANCE ION SOURCES FOR ACCELERATOR MASS SPECTROSCOPY

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Abstract

A project using accelerator mass spectrometry (AMS) is underway at the ATLAS facility to measure the atom densities of transmutation products present in samples irradiated in the Advanced Test Reactor at INL. These atom densities will be used to infer effective actinide neutron capture cross-sections ranging from thorium to californium isotopes different neutron energy spectra relevant to advanced fuel cycles. This project will require the measurement of many samples with high precision and accuracy. The AMS technique at ATLAS is based on production of highly-charged positive ions in an ECRIS followed by injection into a linear accelerator. We use a picosecond laser to ablate the actinide material into the ion source. We expect that the laser ablation technique will have higher efficiency and lower chamber contamination than sputtering or oven evaporation thus reducing 'cross talk' between samples. In addition a multi-sample holder/changer is part of the project to allow for a quick change between samples. The results of off-line ablation tests and first results of a beam generated by the laser coupled to the ECR are discussed as well as the overall project schedule.

INTRODUCTION

Advanced nuclear fuel cycles are currently under evaluation in order to assess their potential to cope with new requirements of radioactive waste minimization, optimization of resource utilization and reduced risk of proliferation. This assessment should account for several key features of the fuel cycle, as of irradiated fuel processing, innovative fuel development and fabrication, waste characterization and disposal. In some cases, the impact of nuclear data and of their associated uncertainties can be crucial in order to assess further exploration. The need for accurate data has been pointed out in recent studies devoted to Generation-IV systems, see e.g. [1]. The very high mass actinides can play a significant role in the feasibility assessment of innovative fuel cycles. As an example, the potential build-up of ^{252}Cf when recycling all transuranics in a light water reactor, leads to increased neutron emissions that could impact the fuel fabrication process [2]. As a consequence, the poorly known nuclear data of higher mass transuranics need to be significantly improved.

At present, most evaluated data files provide some information on these isotopes, but up to now, there has been little emphasis on the quality of these data and very little reliable uncertainty estimations have been provided. This situation is due to the difficulty to make both integral and differential cross section measurements for these isotopes.

The MANTRA (Measurements of Actinides Neutrons Transmission Rates with Accelerator mass spectroscopy) project objectives are to obtain valuable integral information about neutron cross sections for actinides that are important for advanced nuclear fuel cycles. The proposed work takes advantage of two experimental facilities: the neutron irradiation capabilities of the Advanced Test Reactor (ATR) at the Idaho National Laboratory and the Accelerator Mass Spectrometry (AMS) capabilities of the Argonne Tandem Linac Accelerator System (ATLAS) at Argonne National Laboratory [3].

In this paper we will concentrate on the requirements of the AMS program and the novel aspects, namely the laser ablation and the multi-sample changer, that are implemented at the ECR ion source to carry out this research project. The requirements placed on the AMS measurements to be performed at ATLAS are quite challenging. These challenges include high-precision isotope ratio measurements, minimization of cross-talk between samples, efficient use of milligram samples, and the processing of an unprecedented number of samples for a facility as complex as ATLAS. Unique element (Z) identification is desirable, but is not expected to be possible except for specific cases.

The measurement configuration for ATLAS uses the ECR-II ion source [4], significantly modified as discussed below, as the source of ions. After acceleration and deceleration (increasing the accelerator m/q resolution but keeping the ion energy within acceptance range of analytical elements) in the ATLAS linac to approximately 1 MeV/u, the actinide ions of interest are counted in the focal plane of the Fragment Mass Analyzer (FMA) [5].

Small Sample Size and Cross-Talk

A major feature of AMS is the ability to analyze small samples. At ATLAS the AMS activities are focused on samples of a few milligrams. For this project, an added complexity is the need to deal with many small samples. The smaller the samples, the less are the radiological problems associated with handling α-emitting actinides for ATLAS operation. The need to measure many small samples as quickly as possible pushes us to develop efficient sample changing techniques for the ECR source and material delivery techniques which minimize source contamination.

We believe the best approach for this situation is to develop laser ablation for the feeding of sample material into the source. With laser ablation a very small and controllable amount of sample material can be introduced into the source without introduction of extraneous material from the sample holder. Also the angular distribution of ablated material by laser irradiation tends to be strongly peaked around the normal to the surface [6] which is expected to improve the efficiency of capture of ions into the plasma and thereby reduce wall contamination. Finally, the form of the sample material (metal, oxide, etc.) is less critical than with the sputtering or oven technique.

The ECR-II source will also be equipped with a quartz liner. The quartz liner will keep the main body of the source relatively clean of actinides, thus simplifying clean-up. Furthermore, there is some operational evidence that cross talk among samples is reduced. This effect has been observed with other AMS projects at ATLAS. A negative to using a quartz liner is that source performance as measured by charge-state distribution and maximum beam intensity is somewhat reduced. But the beam energy is limited by the bending power of the FMA system and use of high charge state ions is not required. A mass-to-charge ratio of ~8-9 will be quite adequate for these measurements.

LASER PARAMETERS

Laser ablation into an ECR source was first developed at ATLAS [7] and used as a plasma diagnostic tool [8] and has since been used by a number of other labs to explore the coupling of laser produced ions into an ECR source. The technique has not been used routinely for ion production and requires development for this application. The controlled release of materials into the plasma by well-focused laser light will eliminate the significant material buildup often seen in the region of the oven throat or beside the sputter cones, two techniques widely used for sample feeding to the ECRIS. This inefficient, indiscriminate injection of material into the source not only reduces the overall sensitivity of the method but is a major source of cross-contamination between samples. Our experience with lasers in the past indicates that the laser ablation approach will be much cleaner, but must be shown to work for this application.

Laser ablation is a term used to describe removal of material by laser action and it is distinguished from evaporation in equilibrium conditions. In order to remove an atom from a solid by means of a laser pulse one should deliver an amount of energy that exceeds the binding energy of that atom. Therefore the absorbed energy density per atom in a laser-heated layer E_{abs}, should at least be comparable to the heat of vaporisation in equilibrium;

$$E_{abs} = 2AF(t_p)/n_a l_s . \qquad (1)$$

here A is the absorption coefficient; F is the incident laser fluence, the energy per unit surface area during the pulse of duration t_p ; n_a is atomic number density and l_s is the skin depth of the laser in the solid [9]. There are three regimes of laser ablation depending on the laser and target parameters: The thermal ablation, the non-equilibrium semi thermal ablation and the extreme non-equilibrium electrostatic ablation. In the thermal ablation the laser pulse is longer than the major relaxation routes in the irradiated material. Heat conduction and hydrodynamic processes cause the removal of the atoms from the solids. As a consequence the ablation is accompanied by the formation of a large heat- affected zone and throws out molten material [10]. The second regime, non-equilibrium and semi-thermal, is realized when electrons have enough time to transfer the energy to the lattice and the average energy of the ions (temperature) exceeds the binding energy but the distribution function is far from the equilibrium Maxwell distribution. In these conditions, the majority of ions escape the solid before the equilibrium distribution is established. The extreme ablation regime, electrostatic ablation, is completely non-equilibrium and non-thermal. This mode is realised when a short powerful pulse elevates average electron energy during the pulse in excess of the sum of the binding energy of the ions plus the energy necessary for the electron to escape from a solid. The lattice remains cold during the pulse. The energetic electrons escaping from a solid create a huge electrostatic field of charge separation, which pulls the ions out of a solid [11].

In order to avoid a flow of macroparticles molten materials into the ECR ion source, we chose a laser with picoseconds pulse duration. The properties of the laser that we are using in this application are:

- λ =1064 nm
- 15 ps pulse width
- Repetition rate up to 400 Hz
- Pulse energy: variable, up to 5 mJ per pulse.
- Maximum, power of $3x10^8$W per pulse.

OFF LINE TEST SET UP AND RESULTS

For characterization of the laser and to acquire a better understanding of the laser ablated material coupling with the ECR ions source plasma we preformed some test in an off line setup. The Experimental set up is shown in Fig 1. A 4000mm focusing lens is placed at the entrance of a vacuum chamber to mimic the distances required at the ECR source. The result of the optical set up is a focal spot

with a diameter of 450μm at the FWHM. The maximum laser intensity at the focal spot is $2x10^{11}W/cm^2$. An image of the focal spot is presented in Fig 1.

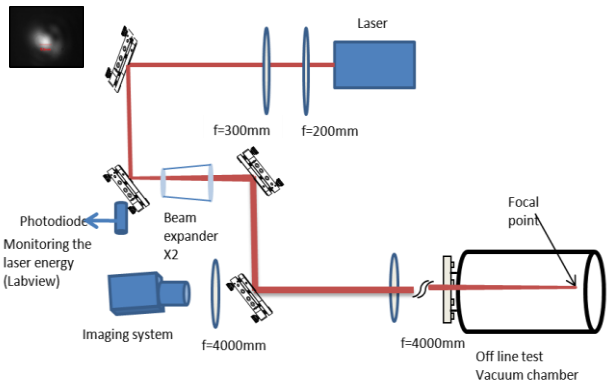


Figure 1: Off line test set up and the laser focal spot image.

The laser induced ablation creates a plasma plume which rapidly expands. Typically the plasma expansion speed is in the order of 10^6 cm/sec. During a picosecond ablation the number of ejected species is about 10^{13}atoms/pulse. The ion flux is about 1% [12]. Using a faraday cup located at a distance of 15.8cm from a solid Ta target we extracted the ion velocity by means of time of flight technique. The calculated averaged ion energy is 40±10 eV. For Ti the corresponding velocity is $1.2x10^6$cm/sec .

A table of examples of ablated material results is included below. In these measurements the laser energy was 1.5-1.6mJ with repetition rate of 400Hz. The resulting laser peak flunce is $0.7J/cm^2$. These studies now provide us with the background information needed to better understand the laser/source performance when the laser is coupled to the ECR source.

INSTALLATION AT THE SOURCE

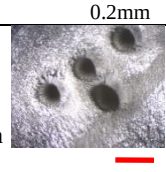
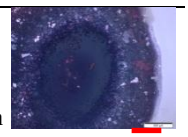
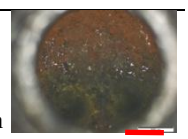
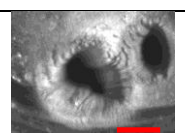
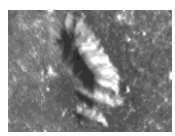
The installation of the laser at the source is shown in Fig. 2. The laser beam is delivered into the source through the extraction aperture. The ablation target is located at the rear of the ECR chamber near the adage of the plasma.

Multisample Changer

Due to the relatively large number of samples (up to 50) to be measured in this experiment and the need to rapidly switch between samples to track any changes in accelerator transmission, an entirely new approach to sample handling, one that is fully automated, is required.

A sample changer that can accept 20 samples has been designed and will be mounted on the injection side of the ECR ion source as shown in Fig. 3. Each sample is mounted onto the end of a 2900mm long aluminum rod that travels in UHV compatible plastic holders. The long longitudinal distance places the mechanism outside of the high magnetic field region of the ECR source. The time to change between samples is less than 1 minute. An absolute encoder is mounted so position information is preserved. In addition, a laser sensor is attached to ensure sample is retracted before rotating. The operation of the multisample changer can be controlled by the accelerator crew or automatically by an experiment program.

Table 1: Ablating rates for different materials

Sample	Consumption rate	Hole depth	Image
Fe solid (1 location shooting for 39 min)	1.3mg/39min 0.033mg/min	1.2mm (for 39 min)	 0.2mm
Fe solid (3 locations 13 minutes on each location)	1.4mg/39min 0.035mg/min $3.7x10^{17}$ atoms/min	1.19mm (for 13 min) 0.09mm/min	 0.5mm
Fe oxide powder-MANTRA target (3 locations 13 minutes on each location)	1.3mg/39min 0.033mg/min $1.24x10^{17}$ atoms/min	1.07mm (for 13 min) 0.08mm/min	 0.2mm
Al oxide powder-MANTRA target (3 locations 10 minutes each)	0.1mg/30min 0.003mg/min $1.77x10^{16}$ atoms/min	0.8mm (for 10 min) 0.08mm/min	 0.5mm
Tb +Fe oxide powder MANTRA target (2 locations 10 minutes each)	0.1mg/20min 0.005mg/min $8.2x10^{15}$ atoms/min	0.57mm (for 10 min) 0.05mm/min	 0.5mm
U metal (3 locations 10 minutes each)	4mg/30min 0.13mg/min $3.289x10^{17}$ atoms/min		 0.2mm
U oxide (3 locations 10 minutes each)	0.5mg/30min 0.016mg/min $3.56x10^{16}$ atoms/min		

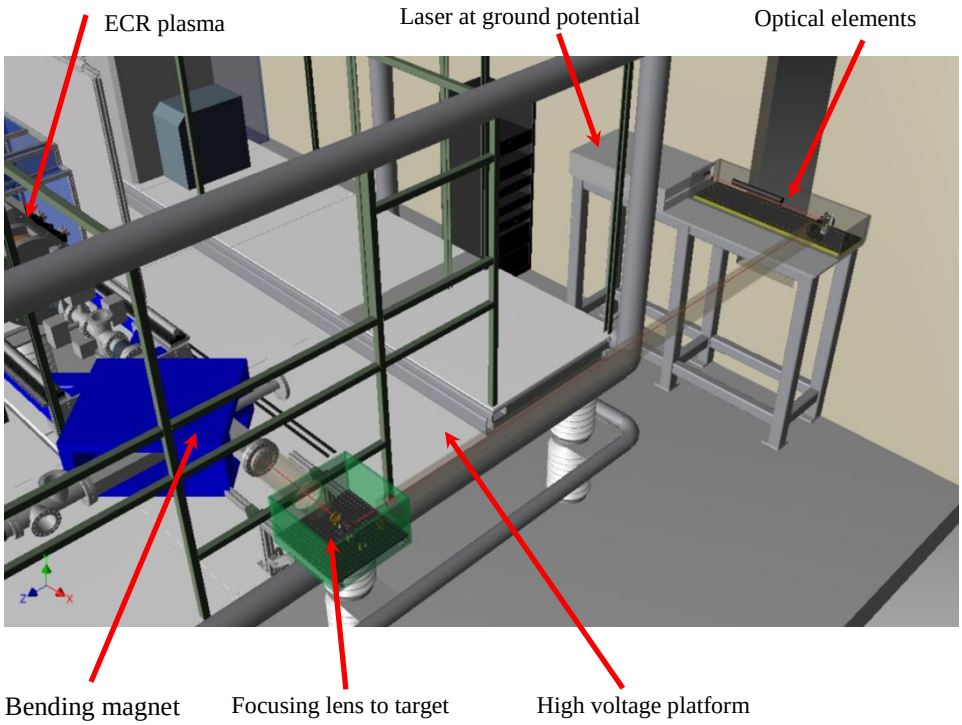


Figure 2: Drawing showing the laser ablation system relative to the ECR source HV platform.

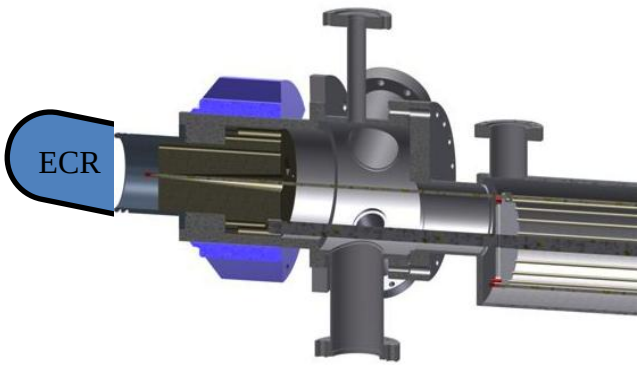


Figure 3: Multisample changer.

Back Light

In order to monitor the laser beam hitting the target samples, we developed an imaging system. The imaging system is located behind the last mirror that directs the laser beam into the source (see Fig 1). We installed a halogen light at the back of the iron taper, (see Fig 4). This way we can collect the scattered light behind the sample holder when it is in place. Using our imaging system we are able to image the outer edges of the sample holder. During a run when the laser hit the sample we are able to identify where the laser hits the sample.

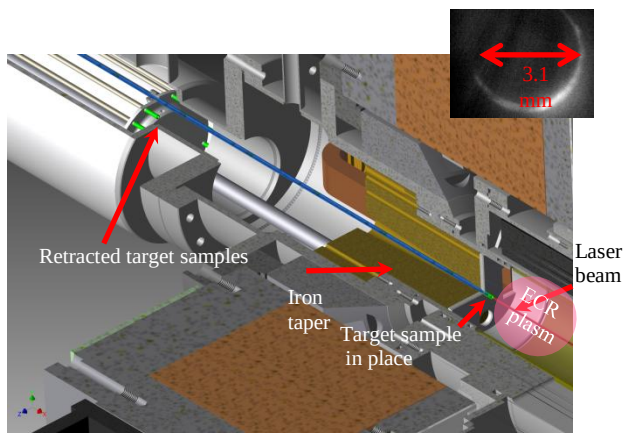


Figure 4: Multisample changer mounted on ECR source showing sample location with the back light.

TITANIUM SAMPLE AT THE ECR SOURCE

Initial tests of the laser coupled to the ECR source took place with a Ti samples. Fig 5 shows a Ti sample after irradiation. The laser parameters that we used during this test are 25Hz repetition rate and a variable energy of 0.5-1.5mj per pulse. The resulting peak intensity is $5x10^{10}w/cm^2$. The overall consumption rate was 0.3mg/hour.

Beam from Ti sample ablated into ECRIS

By means of laser ablation on a Ti sample we were able to generate a high charge state beam. The charge state distribution is shown in Fig 6. From the charge state distribution it is clearly evident the beam production drops to a very low level of 0.1µA when the laser is off.

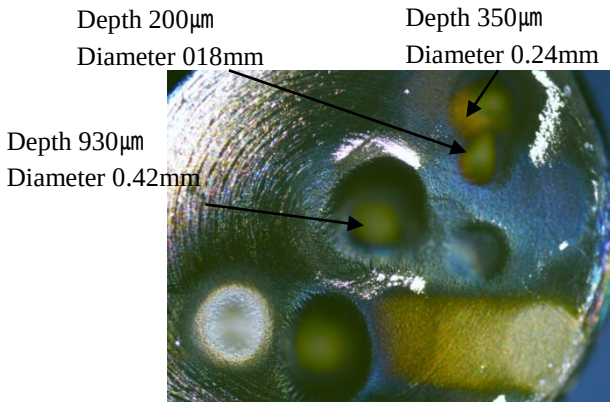


Figure 5: Ti sample after laser irradiations.

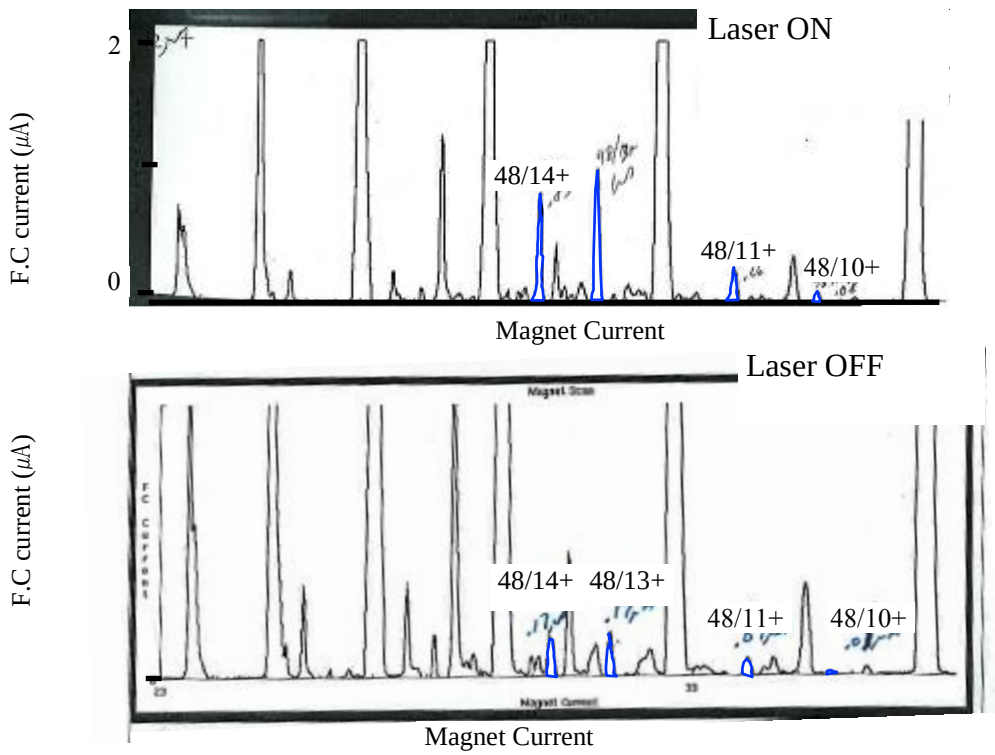


Figure 6: Charge state distribution of Ti beam. The Ti peaks are labeled with mass/charge state in the two figures. The charge–state distribution peaks at 13+. For sputter technique the charge–state distribution peaks at 10+-12+.

Long-term Beam Output from Ablated Ti Sample

For long term stability (100minutes) we measured the 48/13+ charge state. The laser energy that we used for those measurements was 1.5mj with 25Hz repetition rate. The focal beam diameter was 0.5mm. Fig 7 shows the faraday cup current trend with the corresponding laser energy.

The generated beam is around 4 µA and stable for the first 10 minutes and then it drops by 80% for the next 20 minutes while on the same time the laser energy is stable. After that the beam stays stable at a lower level of 0.6 µA for more than an hour.

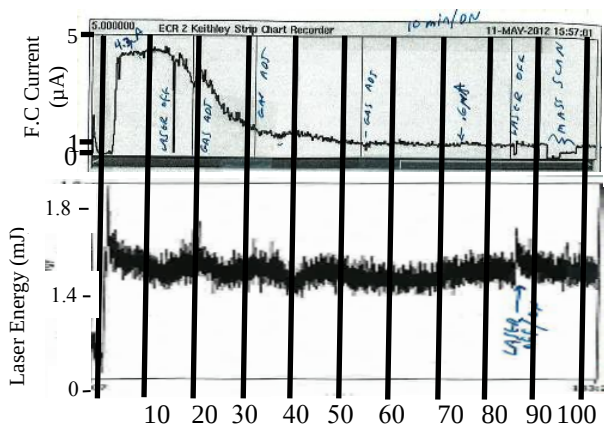


Figure 7: Long term of Ti beam and the corresponding laser energy for 0.5mm diameter in the focal spot.

One of the reasons for this beam current instability current is the drilling rate of the laser in the sample. In order to lower the drilling rate and to gain more stability we changed the laser focal spot on the target to be a bigger. With this new alignment the focal spot was elliptical with major and minor diameter of 0.8mm and 0.6mm respectively.

Fig. 8 shows the long term beam output in the case of the bigger focal spot. The laser energy and repetition rate were the same as in the previous run. Again we measured the 48/13+ charge state. The beam starts with a drop of 37% in the first 2 minutes. After that it stays stable for 20 minutes and then drops 43% in the last 15 minutes. The corresponding laser energy is not stable over the same period of time. We will continue to work to improve the beam stability.

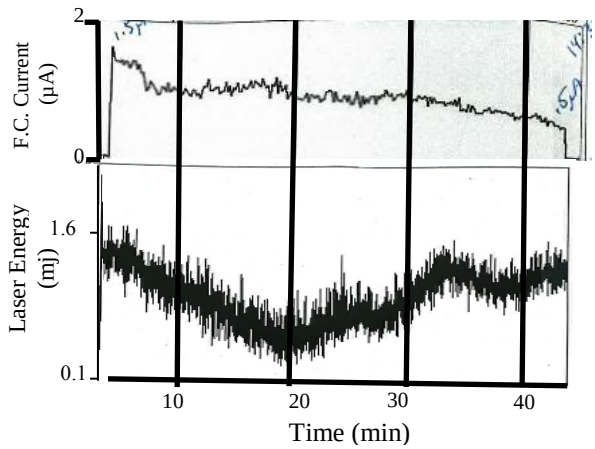


Figure 8: Long term of Ti beam and the corresponding laser energy for the case of elliptical focal spot.

CONCLUSIONS

We had demonstrated high charge state beams generated at the ECR source from laser ablated material. To improve the stability of the generated ion beam we plan to raster continuously the laser beam on the sample. In addition we plan to investigate the influence of the spatial beam profile, especially a hat top profile, on the production of the ion beam. This work is supported by the U.S. Department of Energy, Office of Nuclear Physics, under contract No. DE-AC02-06CH11357.

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