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ULTRASENSITIVE MASS-SPECTROMETER "LIDIA"

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The problems associated with the new island of stability in the region of Z = 110-114 and N = 184, the synthesis of and search for these nuclei in nature were considered in a number of papers (see ref.^{/1/}). It can be assumed that the superheavy element (SHE) concentration in the Solar System matter does not exceed 10^{-14} g/g. This limit follows from the neutron multiplicity measurements of meteorites and the products of processing of some hot brines. The identification of the hypothetic nuclide (primarily its mass assignment) necessitates a search for terrestrial samples containing a higher concentration of SHE and the performance of chemical experiments to concentrate this nuclide.

The requirements of very sensitive and rapid analysis can be satisfied by a mass-spectrometric method involving the direct detection of ions. The identification systems based on tandem generators allowed one to reach a sensitivity of $10^{-10} - 10^{-11}$ at/at in SHE searches (ref.²⁷ and³⁷).

A mass spectrometer consisting of a laser ion source, a vapor or gas target, with the subsequent selection of the ions that have undergone charge exchange, e.g., $3^+ \rightarrow 1^+$, and a multi-stage separation system to lower the background due to rescattered matrix ions can provide the relative sensitivity of 10^{-14} at/at for an about 100 mg sample and analysis duration of 2-3 hours. We shall consider the main units of the proposed mass spectrometer (see the Figure).

To specify some characteristics of the laser ion source a model variant was constructed and tested (ref.^{/4/}). The total number of ions extracted in one laser pulse (we used an YAG laser with Q-spoiling), at the power density of 4×10^9 W/cm², is equal to 1-3x10¹⁰ that is 10^{-3} of the total ion number and 3×10^{-5} of the total number of evaporated atoms. The ion beam emittance, at the accelerating voltage of 40 kV, is equal to 75 mm mrad and, according to estimates, can be decreased by a factor of 5 by using 4-electrode exctraction optics with 100 kV accelerating voltage. The non-selective evaporation and atomic ionization of target material occur at radiation flux density $\geq 5\times10^8$ W/cm², the relative yield of ions depending only on their mass number and being proportional to $A^{-\frac{1}{2}}$. The number of ions with charge states 1^+ , 2^+ , 3^+ successively decreases giving $8 \leq J 1^{+}/J 3^+ \leq 20$.

Owing to the high temperature of the laser plasma, the yield of charged molecules is <u>small as compared with other sources</u>. We observed a weak yield of charged molecules whose energy was

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 $\begin{array}{c} Q2 & 3 & Q3 \\ Q2 & 3 & Q3 \\ D1 \\ 1 & 1^{+} & 2^{+} & 1^{+} & 0^{+} & 3^{+} & 1^{+} \\ 1 & 1^{+} & 2^{+} & 1^{+} & 0^{+} & 3^{+} & 1^{+} \\ \hline 1 & 1^{+} & 2^{+} & 1^{+} & 0^{+} & 0^{+} \\ \hline 1 & 1^{+} & 2^{+} & 1^{+} & 0^{+} & 0^{+} \\ \hline 0 & 1^{+} & 2^{+} & 1^{+} & 0^{+} \\ \hline 0 & 1^{+} & 2^{+} & 1^{+} & 0^{+} \\ \hline 0 & 1^{+} & 2^{+} & 1^{+} & 0^{+} \\ \hline 0 & 1^{+} & 2^{+} & 1^{+} & 0^{+} \\ \hline 1 & 1^{+} & 0^{+} & 0^{+} \\ \hline 1 & 1^$

Diagram of ultrasensitive massspectrometer (LIDIA),

below 2 eV, whereas the energy of atomic ions was equal to 50-600 eV. By selecting ions in energy we succeeded in eliminating charged molecules to 5×10^{-6} of the total ion current, this corresponding to our limit of sensitivity. At the sensitivity level achieved, no yield of charged molecules was observed in the current of doubly and triply charged ions.

In the proposed system we intend to use for analysis only ions having the initial charge state 3⁺. The ions produced in laser plasma are extracted from the ion

source and focused into slit 2. A gas or vapor target and the electrostatic deflector precede this slit. Voltage on deflector plates is chosen so that slit 2 should be struck by the ions with charge state 3^+ at exit from the ion source and converted to be singly-charged after passing through the target. The target thickness of $2x10^{15}at/cm^2$ is sufficient to convert 30-40% of 3^+ -charged ions to the 1^+ - state*. With this thickness, a light target practically does not influence the angular and energy distribution of the beam. A second purpose of the charge exchange cell is an additional decrease of the molecules content in the ion beam due to their dissociation in atomic collisions.

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The ion-optical system of the mass spectrometer is designed on the basis of conventional magnets and lenses to provide the ion mass separation and the lowering to the necessary level of the background due to the scattered matrix ions (elastic, charge exchange, dissociation of molecules). At first we perform the separation of ions with masses lying in the range under study ($\Delta M/M = 5\%$) at a high decontamination factor and then carry out analysis at moderate mass resolution. To achieve the necessary level of preliminary decontamination it is supposed to use a stigmatic achromatic system. The image obtained can serve as a source for a mass spectrometer. By using slits in the places of intermediate images we obtain a multi-stage system with a decontamination factor of 10^6-10^7 per stage.

* This target thickness was obtained from the cross sections measured by us at the charge exchange of 100 keV Ta ions on nitrogen: $\sigma_{0+1+} = 5 \times 10^{-16} \text{ cm}^2$, $\sigma_{0+1+} = 1.2 \times 10^{-14} \text{ cm}^2$.

The part of the ion-optical system consisting of quadrupole lenses Q1-Q4 and dipole magnets D1 and D2 constitutes a system giving the achromatic image of slit 2 in the plane of slit 4. In the median plane (3) this system produces an intermediate image with 1.1 cm/% of mass dispersion. The image obtained on slit 4 serves as a source for the mass analyser consisting of dipole magnet D3 and quadrupole lens Q5. The dispersion in the focal plane of this analyser is equal to 2 cm/ λ . The mass resolution is equal to 500. The radius of curvature of the central trajectory in dipole magnets is equal to 1 m, the bending angle is 90°, the width of magnetic track is equal to 15 cm, the gap is 8 cm. The quadrupole lenses 01-05 have an aperture of about 15 cm and a maximum gradient up to 6 T/M. To reduce to a minimum the main aberrations of the system use is made of correcting elements in the form of the curved boundaries of the dipole magnets. The transmission coefficient of the system is close to 100% for a beam with 20 mm mrad emittance in the horizontal and vertical planes. All calculations for ion-optical properties were done using the standard routine TRANSPORT (ref. /5/).

To achieve a maximum degree of decontamination at each stage the vacuum pumping system should produce an average vacuum not worse than 10^{-8} torr in the entire volume after slit 2 and along thé entire length (about 15 m) of the system at a relatively small (<15 cm) diameter of the chamber. Integrated ion and titanium sublimation pumps are used as an evacuation unit.

The chamber is made from stainless steel, heated to 400 °C and furnished with the necessary set of valves and vacuum sensors. The production of ultrahigh vacuum in the ion source and chargeexchange device is not planned. As was shown by model experiments, the gas flow extracted from the laser-irradiated target with complex composition can reach 10^{-4} torr. ℓ /pulse.Therefore, the ion source and the charge-exchange device will be separated from the rest of the chamber by a differential pumping system. In model experiments a vacuum of 6×10^{-11} torr was produced in a 150ℓ chamber.

It is supposed to use detectors based on channel plates. It is foreseen to employ two detector modules in the following two modes of operation: (a) in the ion current detection mode, at the analysis of the samples for the content of the matrix and trace elements (>10⁻⁶ at/at), and (b) in the mode of counting single ions with time selection, at searches for the SHE and trace elements analysis (<10⁻⁶ at/at).

As the laser ion source operates in the pulsed mode with a current pulse duration of $<10^{-5}$ s, time selection provides the lowering of the background due to the intrinsic background of the detector and scattered ions. For example, at the exposure duration of 10^{6} laser pulses the detection time will be as little as 10^{4} s and the detector background 2 pulses per mass unit. The

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channel plates permit the detection of up to 10^4 ions per laser pulse per mass line. As a result, the dynamical range of the ion currents detected lies up to 5×10^9 .

In conclusion we shall give the estimates of the highest sensitivity that can be achieved by this system. Taking into account the yield of triply-charged ions and the losses due to charge exchange, we obtain 10⁹ ions per l'aser pulse. At the pulse frequency of 100 Hz about 10^{15} ions can be extracted through slit 2 for 3 hours. In this case the material consumption will be 3×10^{19} atoms or 10 mg as recalculated for Pb.During this period of time, 10 ions of the sought nuclide will arrive at the detector device if its concentration in the sample is equal to 10⁻¹⁴ at/at. The actual sensitivity can turn out to be somewhat worse due to residual charged molecules, the background and a less than 100% detector efficiency. Even if it is equal to 10¹² at/at, the mass spectrometer will be useful in searches for SHE. Its advantage is that it allows both a search in the mass range $A/\Delta A = 20$ and accurate mass number identification at a resolution of about 500.

REFERENCES

- Flerov G.N., Ter-Akopian G.M. Pure Appl.Chem., 1981, 53, p.909-923.
- Stephens W., Klein J., Zurmühle E. Phys.Rev., 1980, C21, p.1664-1666.
- 3. Schwarzschild A.Z., Thieberger P., Cumming J.B. Bull.Am. Phys.Soc., 1978, 22, p.94.
- 4. Arzumanian G.M. et al. JINR, P7-81-744, Dubna, 1981.
- 5. Brown K.K., Kear B.K., Howry W.K. Report SLAC No.91, Stanford, 1970.

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Описан высокочувствительный масс-спектрометр с лазерным источником ионов для поиска сверхтяжелых элементов /A>250/.Показано, что можно создать прибор с относительной и абсолютной чувствительностью 10⁻¹²-10⁻¹⁴ат/ат и 10⁶ ат соответственно, получаемой при времени анализа продолжительностью около трех часов.

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Ter-Akopian G.M. Ultrasensitive Mass-Spectrometer "LIDIA"

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An ultrasensitive mass spectrometer using a laser ion source has been designed to search for superheavy elements (A> 250). It has been shown that it is possible to build a device with relative and absolute sensitivities of respectively $10^{-12} - 10^{-14}$ at/at and $\cdot 10^{6}$ atoms for analysis duration of 3 hours.

The investigation has been performed at the Laboratory of Nuclear Reactions, JINR.

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