## **Multiply Charged Ions and Their Effective Applications**

Victor V. Apollonov, Sergei M. Sil`nov

Prokhorov GPI RAS, Vavilov str.38, 119991, Moscow, Russia

## Abstract

The creation of high-power lasers has opened a new era in the development of basic research and cuttingedge technologies in various fields of practical application. All this is, first of all, due to the unique properties of laser sources of high-power coherent radiation. They include: a) high monochromaticity (i.e. a small width of the emission line), which offers new opportunities in high-resolution spectroscopy; b) high spatial and temporal coherence (i.e. the occurrence of light oscillations in a coordinated manner, resulting thereby in a distinct interference pattern), which gives a strong impetus to the development of holography and optical information processing methods; c) a relatively high specific energy that can be emitted by the laser, d) a possibility of varying the length of time during which the energy stored in the laser can be emitted in a wide range of durations: from continuous to femtoseconds; e) a possibility of variations in the temporal structure of radiation from tens of Hz up to tens of GHz; and f) a small divergence, which enables tight focusing. Due to these properties, the laser power density, which can affect the substance, reaches a giant value on the order of 10<sup>20</sup> W/cm<sup>-2</sup>. Consequently, it is possible to expose a substance to radiation whose power density exceeds all known today values characterizing natural and artificial sources. This fantastic opportunity has been thoroughly investigated in the recent decades by scientists from different countries. Clearly, by gradually increasing the laser energy and reducing the length of time during which that energy is emitted, it is possible to observe several stages of an interaction like this.



**Figure 1**. Scheme of a laser-plasma ion source: *1* – laser, *2* – focusing lens, *3* – target, *4* – laser plasma, *5* – ion beam having a maximum charge and maximum energy.

Thus, we first observe a gradual increase in temperature, which will result in heating of almost any substance up to the melting temperature. Then, a further increase in the rate of energy input into the material leads to such a rapid heating that an intensive evaporation starts, bypassing the liquid phase. Under these interaction

conditions, one can observe a phenomenon, which seems surprising at first glance: with increasing power density the amount of the evaporated mass of the matter remains virtually unchanged. The explanation of this phenomenon is simple enough. It consists in the fact that as a result of intense evaporation, the material's vapor begins to screen the irradiated surface. Therefore, a significant portion of the laser energy no longer reaches the surface and is absorbed in the vapor produced by laser radiation. In this case, with increasing flux density of the laser energy the vapor temperature should increase, which will lead to the formation of plasma, i.e., ionization of the evaporated material and creation of a plasma screen, preventing a further input of energy into matter. This fact is not surprising and, moreover, extremely high vapor temperatures, up to strong ionization of evaporated material atoms, can be achieved. The temperatures reached due to laser irradiation were so high that scientists had a desire to try to realize the fusion of light nuclei, i.e., to implement a fusion reaction on the scale of a pinhead.

The idea of laser fusion, associated with the ability to create conditions for thermonuclear reactions in the area of laser radiation focusing on a solid target, gave birth to a new area of research, i.e., physics of the interaction of intense laser pulses with matter, whereas the plasma obtained by this method was called "laser plasma". This new direction has progressed dramatically for the last 40 years due to the rapid development of laser physics and engineering. Long-term efforts on the experimental and theoretical study of the properties and characteristics of laser plasma have extended the range of effective applications of laser plasma, originally associated only with laser thermonuclear fusion.

Instead of laser radiation one can also use high-energy particle beams, which provides a significantly more efficient transmission of the deuterium-tritium fuel energy. A beam of heavy multiply charged ions seems the most promising in this case, because particles with a high charge can be easily accelerated to high energies. Besides, it is easier to obtain higher current values, since the greater the mass, the smaller the influence of the repulsion of ions in the beam due to a space charge. In addition, the beams of heavy multiply charged ions will transfer energy to a target with maximum efficiency. In this case, there arises a question of how to obtain primary beams of heavy multiply charged ions for subsequent acceleration to high energies in accelerator systems. From this perspective, a simple way to implement an inertial controlled thermonuclear reaction is first to obtain the beams of heavy multiply charged ions with the help of high-power laser radiation and then to use these beams for irradiating a deuterium-tritium target in order to reach the fusion temperature. Thus, the methods of obtaining high temperatures and a highly ionized state are of great practical interest.

The main parameters of an ion source are the charge-state distribution of the ions produced and the intensity of the extracted ion beams Most of the currently existing types of multiply charged ion sources are based on subsequent electron-impact ionization. By 1970s heavy-ion accelerators had mainly relied on the use of an ion source based on a reflective (Penning) discharge with oscillating electrons. Ion sources like these exhibited a theoretical limit of charged particles and the intensity of extracted beams, which is determined by the temperature, density and lifetime of the plasma. The electron-beam method of multiply charge ion production was proposed by E.D. Donets in 1967. This ion source, later called an electron-beam ion source (EBIS), was conceived as a pulsed source to produce multiply charged ions for high-energy ion accelerators. To increase the charge of the ion produced by an EBIS, the energy of ionizing electrons should be equal to 100 - 200 keV, and the retention time of the ions should amount to tens of seconds or even minutes. The EBIS uses a linear electron accelerator operating in the continuous mode. Ion sources with a short electron beam having a length of less than 10 cm formed a separate line of research and were called an electron-beam ion trap (EBIT). During the 1970s and 1980s the EBIS's were used to produce record-high charge states of heavy ions. Using a KRION-2 cryogenic electron-beam ionizer, scientists at the Joint Institute for Nuclear Research (JINR) (Dubna, Russia) managed to produce highly charged xenon ions, while using an EBIT, researches at the Lawrence Livermore National Laboratory (LLNL) (Livermore, USA) produced highly charged uranium ions. The main disadvantages of the EBIS in comparison with other types of sources are ion losses and relatively low intensity (10<sup>10</sup> s<sup>-1</sup>) of the produced ions. The most widely used sources of multiply charged ions for accelerators and nuclear physics are electron-cyclotron resonance (ECR) sources. An ECR source is an open magnetic trap for plasma confinement. Electrons and ions are generated as a result of electron impact ionization. In turn, the

electrons resulting from the ionization of neutrals and ions are heated to an energy of several keV by the microwave radio-frequency field, whose frequency is equal to the Larmor frequency of the electron spin in the longitudinal magnetic field of the trap. Increasing the degree of ionization in the ion source is the result of successive ionization during the holding period of the ions. To date, all large accelerator centers in the world are equipped with such ion sources. However, modern requirements to the sources significantly exceed their capabilities. The first step is to increase the ion beam current because ESR sources are promising for continuous ion accelerators due to their ability to generate rather intense (up to 10<sup>13</sup> s<sup>-1</sup>) beams of medium and heavy ions in continuous mode.

In contrast, a laser-plasma generator of multiply charged ions produces a large number of heavy ions in the mode of short periodic pulses, which is of interest for ion accelerators operating in pulsed mode. The source of this type is also promising for research in the field of heavy-ion fusion. The principle idea of a laser-plasma source of multiply charged ions was proposed 45 years ago together with author of the book by researchers at the Moscow Engineering Physics Institute (MEPhI). The laser-plasma generator is based on the physical phenomenon of generation of highly ionized states of atoms under irradiation of the surface a solid target by a high-power, focused laser pulse [1]. When expanding into a vacuum, high-temperature laser plasma produces a powerful stream of charged particles. The advantages of a laser-plasma source as compared to other types of ion-pulse generators include:

- ability to generate multiply charged ions of almost any elements of the periodic table, and
- the ability to generate an intense, short  $(1 100 \ \mu s)$  ion pulse with a record-high brightness.

The main difference of the laser plasma from other high-temperature plasma objects is a high energy density in matter, caused by the ability of lasers to concentrate in a short time the energy in small volumes of material (10<sup>-6</sup> cm<sup>-3</sup>). Changing the power and wavelength of heating radiation allows one to control the temperature and the laser plasma density. Irradiation of a target surface by focused laser radiation with a power density  $>10^8$  W/cm<sup>2</sup> produces a plasma plume with a large temperature and density. Depending on the laser radiation parameters, the electron temperature in the plume can be as high as  $T_e = 2.10$  keV at an electron density of  $10^{19} - 10^{21}$  cm<sup>-3</sup>. This allows one to produce highly charged states of ions in the laser plasma. Such characteristics of the ion component of the laser plasma as highly charged states, intensity and type of angular distribution are exceptionally favorable for its use as a source of multiply charged ions for accelerators. The first laser-plasma generator for an accelerator complex was constructed thirty years ago (joint work of MEPhI and JINR researchers). Requirements imposed by accelerators to ion sources (pulse repetition rate, up to 10 Hz; stability of the output parameters of the ion beam,  $\pm$  10%; and time of continuous operation) set strict limits as to the choice of the laser type and its maximum achievable energy. Due to the development of laser technology in the last 30 years, transverse discharge pumped CO<sub>2</sub> lasers seem most attractive for the use in laser-plasma generators. The output energy of these lasers can vary from 1 to 1000 J at laser pulse durations from 0.01 µs to 1 µs and a pulse repetition rate up to 10 Hz. Using CO<sub>2</sub> lasers to generate multiply charged ions from laser plasma, due to their high level of technical development, relative ease and low cost even in the configuration of a low-frequency repetitively pulsed facility with a long running time, is currently very promising. An increase in the repetition rate of high-power and short laser pulses up to 30 - 50 kHz will significantly increase the yield of multiply charged ions and dramatically enhance the efficiency of laserplasma generators.

Thus, evaporation of the material and heating of the vapor lead to a plasma state, i.e., when vapor is heated, electrons are detached from atoms due to a temperature rise. In this case, the higher the temperature, the more electrons are 'torn away' from an atom. Therefore, an ionized vapor, i.e., a plasma, consists of electrons and ions. If we assume that the temperature of the electrons and ions is the same, and since the electrons are much lighter than the ions, they travel much faster. Due to such a high velocity, the electrons in the process of the gas-dynamic expansion will be the first to fly out of the plasma region. Then, at the forefront of expanding plasma there occurs separation of the negative charge of the emitted electrons and the positive charge of the

ions. In this case, due to the Coulomb interaction, the electrons pull the ions. This process will lead to the separation of electrons and ions in space and time (quenching effect), which interferes with the recombination of electrons and ions in a laser plasma. Therefore, a directional flow of highly charged ions is produced in the form of a beam that propagates in a direction normal to the target surface. For ions with a maximum charge and energy, the angle of their departure decreases, and one can observe the effect of self-focusing of ions, depending on the charge multiplicity. In this case, the interaction of a focused laser beam with a high energy flux density gives us a very simple and effective source of multiply charged ions without the need for any pulling or focusing fields.

Fig. 1 shows a scheme of a laser-plasma source. In this laser-plasma source of ions, the ionization degree is controlled by the power density of laser radiation and can reach a few dozen; this means that it is possible to completely ionize a significant part of the elements of the periodic table. It should be noted that prior to the advent of lasers it was practically impossible to achieve the degree of ionization Z>10 and thus, for example, to measure the bonding energy of the electrons in the atom. We emphasize that in this case new practical and promising applications, rather than purely fundamental results, proved to be important.

A laser-plasma source of ions and nuclei is promising for charged particle accelerators. In January 1976, MEPhI and JINR scientists realized for the first time in the world the acceleration of carbon nuclei from the laser-plasma source (Fig. 2). Carbon nuclei from the laser-plasma source were introduced into a linear accelerator and accelerated in the synchrotron channel to energy of 50 GeV. If previously the accelerator accelerated singly charged protons, the use of multiply charged ions made immediately increased the energy of accelerated particles by Z times. This made it possible to make the next step in obtaining relativistic beams of complex nuclei. In recent years, such work has been carried out at CERN (Switzerland) to obtain multiply charged ions of heavy atoms with Z>50 by using a laser-plasma source of ions.



**Figure 2**. Scheme of the acceleration of carbon nuclei at the proton synchrotron using a laser-plasma source of carbon nuclei  $C^{6+}$ : 1 – laser, 2 – lens, 3 – graphite target, 4 – linear accelerator, 5 – synchrotron, 6 – output beam.

Nowadays, laser plasma finds numerous applications in various fields of fundamental physics: X-ray spectral analysis of multiply charged ions, high energy density physics and physics of shock waves, modeling of space-physical and high-temperature processes, X-ray lithography, etc. In recent decades, ion sources have been intensively designed and developed. An incentive to create them stems from the need of heavy-ion accelerators. The ion source is the first element of an accelerating complex, which determines the structural characteristics, parameters, efficiency and capabilities of the accelerator. Generation of high-current beams of

multiply charged ions is one of the most promising areas of research, which finds wide application in science and technology. As an example of employment of such beams, we can single out the following:

- a source of multiply charged ions of heavy elements in the fore-injectors of particle accelerators;

- a source of cluster ions and molecules;

- a source of beams of highly ionized atoms to measure the interaction cross sections in such field of atomic physics as nuclear fusion, ionosphere physics, astrophysics and investigation of the laser plasma;

- production of uniform films to form the fine structure of multilayer X-ray mirrors and diamond films;

- radiation material science, ion implantation to alloy semiconductors or change the surface properties of materials; and

- formation of ion beams with a specific charge state in order to create a gain medium for the X-ray lasers.

Recent years have seen a significantly growing interest in a wide practical use of sources of multiply charged heavy ions. Thus, in Dubna (JINR) scientists have developed a unique technology for producing filters. In a polymer film a passing heavy ion leaves a channel having large radiation damage, and complex molecules are broken down into smaller parts, i.e., radicals. For this reason, the region of the track (channel) becomes sensitive to chemical reactions. After etching the polymer film exposed to heavy ions, through channels are formed. The diameter of these channels depends on the temperature and etching time, and can range from five nanometers to tens of microns. Such channels can be used in biology and medicine for separation of different types of viruses and bacteria (having dimensions greater than 0.2  $\mu$ m), and the protein molecules (enzymes); in addition, they can be used in semiconductor technology and for water filtration.

Heavy-ion beams are quite promising for radiotherapy, in particular for the treatment of cancers. The use of laser-plasma beams of heavy ions is much more effective in comparison with the use of large and expensive installations, because they make possible the radiotherapy of limited areas of the human body due to the large ion charge. Heavy ion sources are also needed for the synthesis of transuranic chemical elements, lying on the so-called 'islets' of stability, which are predicted in the region Z = 114 and Z = 126.

No less interesting and practically important application of laser-plasma ion sources is ion implantation of metals. It has been shown experimentally that the bombardment of the surface of metals by ions can significantly change their physical properties. The reason for this is, firstly, the effect of the introduction of these or other ions, that is, the change of the elemental composition by doping, and secondly, the change in the structure of the surface layers. Such methods have been developed since the 1970s; they have shown that ion implantation can significantly alter the physical properties of metals, including corrosion resistance, surface friction, wear resistance, etc. It should be emphasized that the laser-plasma implanter for modification of metal surfaces is attractive due to the fact that it has a small size and makes it very easy to use this or that type of ions and to obtain multi-element ion beams of required stoichiometric composition. Besides, one can easily obtain a pulse current of the order of an ampere and, using a standard frequency laser, have reasonable time values to ensure the required irradiation dose.

The interaction of laser radiation with matter is another very important and promising sphere of analytical research, whose aim is to determine the elemental composition of the material irradiated by the laser. This physical prerequisite of this phenomenon were the following experimental facts: the possibility of evaporation of any material regardless of its composition, nature and physicochemical properties, i.e., laser radiation at appropriate energy and power parameters can evaporate metals, semiconductors, textiles, dielectrics, ceramics, biological object (living tissue, blood), etc. And, as mentioned previously, due to the plasma shielding an almost constant quantity of substance (about one microgram) is evaporated. The next important factor is that the evaporated material is almost completely ionized in the same laser beam, and the ions are

formed into a beam directed along the normal to the laser-irradiated targets. Thus, a laser-plasma ion source also allows registration and separation of particles in electric and magnetic fields.



**Figure 3**. Scheme of a time-of-flight mass spectrometer with a laser-plasma ion source: 1 - laser, 2 - irradiated target, 3 - electrostatic analyzer, 4 - drift length, 5 - registration system of ions, 6 - oscilloscope.

Fig. 3 shows the scheme of a time-of-flight mass analyzer using a laser-plasma ion source. The simplest scheme involves a combination of laser-plasma ion source with an electrostatic analyzer and time-of-flight mass separation. Fig. 3 shows the principle of operation of the laser device for express elemental analysis. Radiation from the laser is focused on a target, the elemental composition of which we wish to identify. Upon arrival of the laser pulse, a part of the target is vaporized and ionized. The main portion of the ions is formed into an ion beam which is directed along the normal to the surface of the target. In this case, the ion beam corresponds to the target composition. The electrostatic analyzer identifies the ion beam, ideally with a single energy value. Then, this monochromatic beam of ions, formed from elements with different masses, will be separated in time, i.e., the lightest ions will first reach the registration system. From the registration system the electrical signal is fed to an oscilloscope where a certain maximum weight of a chemical element corresponds to each maximum. This time-of-flight mass spectrometer with a laser-plasma ion source allows one to analyze in real time a wide class of materials with a sensitivity of  $10^{-3} - 10^{-4}$  atomic percent with the mass resolution of ~300.

Very useful and promising was the use of a laser-plasma ion source in the well-known scheme of a spectrometer with double focusing, which combines an electrostatic analyzer and a mass spectrometer sweeping the magnetic field. In this case, the ion beam from the target being irradiated passes through the electrostatic analyzer, where the beam is monochromatized and then falls into a uniform magnetic field, where, as is known, ions are separated according to their charge-to-mass ratio. Because use is made of singly charged ions, the beam of ions with different masses is swept by the magnetic field and recorded, for example, on a photographic plate. Such a device exhibits high analytical characteristics. The mass resolution reaches the value of up to 5000, which means that it is possible to detect the isotopes of all elements of the periodic table and register all the elements from hydrogen to uranium. This device also makes it possible to detect impurities in the material, when the concentration is about  $10^{-5} - 10^{-6}$  at.%.

A distinguishing feature of the operation of a mass spectrometer with a laser-plasma ion source, except for its versatility and analysis of any substance, consists in the fact that all the elements are registered at

simultaneously, and knowing the evaporated mass one can perform an analysis without any standards, if use is made of natural isotope ratios of different elements. These characteristics determine the possible applications of mass spectrometers with laser-plasma ion sources, which include not only the control of impurities in metals and semiconductors, but also opportunities for environmental pollution control in the air and in the soil. Thus, when measuring the composition of wheat grains collected closer than 300 meters from heavily loaded transport highways, lead is found. Besides, in pearl lipsticks mercury is detected. Analysis of the scales of fish caught in the water near untreated sewage into the river, a large set of heavy metals is found.

Using such devices in biology and medicine also undoubtedly holds promise for medicine and forensics. Thus, it is possible to carry out an analysis of the elemental composition of blood using a piece of gauze soaked with blood or a substrate with a drop of blood on it [2]. For example, to forensics, it is important that the content of heavy elements in a person's blood is strictly individual [3]. In analyzing biological tissue elements one can detect drastic changes in the number of different elements such as Ca, Cl, Si, Al, etc., which indicates certain diseases.

One cannot but mention one more very important and interesting application of laser plasma. Because a high degree of ionization can be achieved in laser plasma, at the early stages of the expansion the laser plasma becomes an intense source of soft X-rays that are of bremsstrahlung and recombination nature; therefore, we deal with the radiation arising due to changes in the velocity during the interaction of charged particles and the recombination of electrons and ions [4]. In this case, the wavelength of the recombination radiation is determined by the degree ionization and can be controlled both by the laser parameters and by using materials with different ionization potentials. Soft X-ray radiation can be focused and thereby beams with high energy characteristics can be generated.

Such a laser-plasma source is very promising for the use in X-ray lithography in the manufacture of semiconductor chips. Due to the small wavelength and small focusing spot size one can significantly improve the resolution of the photomask used in the production of semiconductor chips. In addition, a laser-plasma soft X-ray source enables the creation of a microscope working in the soft X-ray range, i.e., using photons with such a low energy that will not cause genetic mutations and destroy the structure of a living cell, but will allow one to follow the behavior of individual chemical elements.

Another very promising technological application of laser plasma is the production of thin films of complex composition. In this case, use is made of the flows of the plasma produced in the interaction of matter with radiation of a *Q*-switched laser, i.e., in the mode of high-rate energy input into the matter. Thus the target can be made of different materials determining the desired stoichiometry of the resultant film. In this case, high rates of energy input, as noted above, allow one to preserve the stoichiometric composition of the target and to produce complex compounds on the substrate. The use of frequency lasers makes it possible to deposit materials epitaxially, i.e., in small portions, layer by layer, while maintaining the crystalline structure. In addition to producing complex semiconductor compounds, this technology has been very useful in creating high-temperature multicomponent superconductor compounds.

This technology is characterized by the fact that complex compounds in the form of single crystals grow on substrates. We can assume that one of the reasons for this is the concomitant recombination radiation from the laser plasma, which activates electron shells of atoms during crystallization, which stimulates the growth of crystals with more energy-intensive orientations. Experimentally, many laboratories have shown that by vaporizing carbon by laser radiation from a graphite target, it is possible to produce thin diamond-like films and more complex structures in the condensation process under certain conditions.

Note in conclusion that the sphere of possible applications of multiply charged ions produced in the interaction of high-power pulsed and repetitively pulsed laser radiation with matter is expanding every day. The next step is the introduction of high-power repetitively pulsed laser systems with high pulse repetition rates, providing a solution to the problem of a plasma screen and making it possible to deliver much higher

average power flows into the matter, and thereby producing more powerful beams of multiply charged ions for their effective use [5].

## References

- V.V. Apollonov, Yu.A. Bykovskii, N.N. Degtyarenko, V.F. Elesin, Yu.P. Kozyrev, S.M. Sil'nov. Production of multiply charged ions by interaction between a powerful laser pulse and a solid. *Pis'ma Zh. Eksp. Teor. Fiz.*, **11**, 377 (1970) [*JETP Lett.*, **11**, 252 (1970)].
- 2. V.V. Apollonov, Yu.A. Bykovskii, Yu.P. Kozyrev, S.M. Sil'nov, T.N. Filinskii. Possibility of using mass spectrometry for elemental analysis of blood, Report No. 15/17-75, Moscow Engineering Physics Institute, Moscow, 1970.
- 3. V.V. Apollonov, Yu.A. Bykovskii, Yu.P. Kozyrev, S.M. Sil'nov, T.N. Filinskii. Possibility of using mass spectrometry in forensic science, Report No. 15/17-73, No. 15/17-74, No. 15/17-76, Moscow Engineering Physics Institute, Moscow, 1970.
- 4. V.V. Apollonov, K.Kh. Kazakov, N.V. Pletnyev, V.R. Sorochenko. Current measurements in the plasma produced by train of 10.6 μm sub-nanosecond laser pulses. *Proc. SPIE*, **3886**, 198 (2000).
- 5. Victor V. Apollonov, Sergei M. Sil`nov. Particles Acceleration Processes in the Laser Plasma. International Journal of Advanced Research in Physical Science (IJARPS) *Volume 4, PP 1-14 ISSN No.* (2017)