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High intensity production of high and medium charge state uranium and other heavy ion beams with VENUS^{a)}

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The next generation, superconducting ECR ion source VENUS (Versatile ECR ion source for NUclear Science) started operation with 28 GHz microwave heating in 2004. Since then it has produced world record ion beam intensities. For example, 2850 eμA of O⁶⁺, 200 eμA of U³³⁺ or U³⁴⁺, and in respect to high charge state ions, 1 eμA of Ar¹⁸⁺, 270 eμA of Ar¹⁶⁺, 28 eμA of Xe³⁵⁺ and 4.9 eμA of U⁴⁷⁺ have been produced. A brief overview of the latest developments leading to these record intensities is given and the production of high intensity uranium beams is discussed in more detail.

1. Introduction

Third generation ECR ion sources are designed around operating frequencies of 28 GHz for which 10 kW CW gyrotrons are commercially available. The first 28 GHz heating of an ECR ion source was achieved by the Grenoble/Catania collaboration, which performed a series of tests coupling 28 GHz into the superconducting ECR ion source SERSE^[1,2]. These tests provided important data for the future design of an optimized 28 GHz ECR ion source. The VENUS (Versatile ECR ion source for NUclear Science) ECR ion source, which started its commissioning at 28 GHz a few years later at Lawrence Berkeley National Laboratory (LBNL), is the first third generation source routinely operated at 28 GHz microwave heating. As first high field 28 GHz ECR ion source a number of the technical challenges with respect to the superconducting magnet design and construction, cryogenics and bremsstrahlung heating were encountered. The solutions developed^[3-5] are now being incorporated in other superconducting ECR ion sources under design or construction^[6-8].

One of the key features of the VENUS ECR ion source is its strong magnetic confinement produced by its robust and reliably operating NbTi superconducting magnets. The axial magnetic confinement fields of 4 T at injection and 3T at extraction and a radial field of up to 2.1 Tesla (slightly more than twice the resonant field for 28 GHz ($B_{\text{ECR } 28 \text{ GHz}}=1\text{T}$)) at the plasma chamber wall allow for optimum operation at 28 GHz. The axial field profile is generated by three solenoid coils, which

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allows for a wide range of minimum B fields (B_{\min}) between the magnetic mirror peaks. Besides 28 GHz, 18 GHz can be injected as a second frequency for double frequency heating or for single frequency heating at 18 GHz ($B_{\text{ECR } 18 \text{ GHz}}=0.64\text{T}$) only. Another key feature is the incorporation of a tantalum x-ray shield into the plasma chamber^[4], which reduces the bremsstrahlung heat load to the cryostat. This enables VENUS to be operated at higher powers and to explore different magnetic confinement regimes. However, the bremsstrahlung absorbed into the cold mass and its consequently heating of the cryostat remains a challenge and a limit for the VENUS ECR ion source and will be also a major challenge for 4th generation ECR ion sources^[9]. The VENUS development is driven by two main objectives. First, VENUS serves as the third injector source for the LBNL 88-Inch Cyclotron, boosting both the energy and intensity of beams available from the facility. The primary goals for VENUS as cyclotron injector are first to extend the 5MeV/nuc energy range of the K140 cyclotron all the way to uranium for nuclear structure studies and secondly to extend the 16 MeV/nuc heavy ion cocktail for the BASE (Berkeley Accelerator Space Effects) facility to xenon beams^[10]. These goals require the production of several μA of U^{46+} or U^{47+} and the production of several tens of enA of $\text{Xe}^{42+ \text{ to } 44+}$. After VENUS had been coupled to the 88-Inch Cyclotrons in fall of 2006, a $^{129}\text{Xe}^{42+}$ beam at 14 MeV/nucleon and a $^{238}\text{U}^{47+}$ beam at 4.5 MeV/nucleon were successfully accelerated and extracted from the cyclotron.

The second objective for the VENUS project is the development of a prototype injector source for a high intensity heavy ion beam driver linac to meet intensity requirements of a future rare isotope facility (FRIB) in the US^[11]. The production of intense beams of medium to low charge states ions, in particular uranium beams, are key to success for this and next generation radioactive ion beam facilities such as the radioactive ion beam factory in RIKEN in Japan^[7] and the FAIR facility at GSI in Germany^[8]. Therefore, uranium beam production is a focus of research at LBNL. Extensive oven and material research and development (R&D) has been conducted to produce uranium vapor and intense uranium ion beams. Results of this work will be presented in this paper.

2. Recent beam experiments with the VENUS ECR ion source

Since the commissioning at 28 GHz in 2004, many record ion beams have been produced with VENUS^[3,12]. Recently, we have performed studies with Ar, Kr, Xe and uranium beams and the intensities achieved are summarized in Table 1. For comparison, published results from other high performance ECR ion sources are listed as reference^[13,14]. The ion source performance is continuing to improve as we are

coupling more power into the plasma chamber.

Table 1. Commissioning results for VENUS in comparison with other high performance ECRIS

f(GHz)	VENUS		GTS ⁽¹⁴⁾ SECRA ^{L(13)}	
		28 or 18 + 28	18	18GHz
¹⁶ O	6 ⁺	2850	1950	2300
	7 ⁺	850		810
⁴⁰ Ar	12 ⁺	860	380	510
	14 ⁺	514	174	270
	16 ⁺	270	50	73
	17 ⁺	36	4.2	8.5
	18 ⁺	1		.2
⁸⁴ Kr	25 ⁺	223		
	27 ⁺	85		
	28 ⁺	25		
	29 ⁺	5		
	30 ⁺	1		
¹²⁹ Xe	28 ⁺	222	120	
	29 ⁺	168	*	
	30 ⁺	116	60	101
	31 ⁺	67	40	68
	34 ⁺	41	8	21
	35 ⁺	28		12

Two main magnetic confinement and heating configurations are typically used in the VENUS ECR ion source. In the single frequency heated plasma mode a minimum B field of .64 to .75 T is used, which results in a shallow magnetic field gradient at the 28 GHz resonance zone. Up to 6.5 kW of 28 GHz power has been coupled into VENUS using this mode of operation. In the double frequency mode a minimum B field of .45 T is used. This field profile results in a combination of a shallow gradient (for 18 GHz heating) and a steep gradient (for 28 GHz heating) at the resonance zone. Up to 9kW of combined 18 and 28 GHz power (a power density of about 1kW/liter) has been coupled into the VENUS plasma chamber so far. Figure 1 shows typical axial magnetic field configuration used for the VENUS ECR ion source for the two modes. The sextupole magnet is energized to produce 2 Tesla at the plasma chamber wall in all cases.

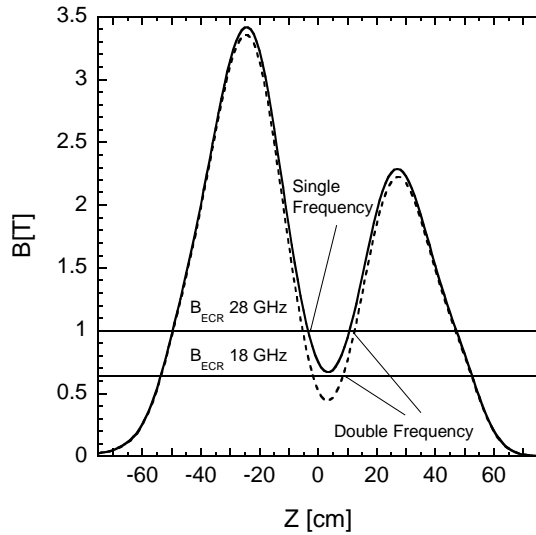


Fig.1 Typical axial field profiles used for the single frequency heating at 28 GHz and double frequency

3. Development of high intensity uranium beams

High intensity uranium beams are especially challenging to produce because of the chemical properties of uranium and the high temperature required to evaporate enough feeding material for the plasma. In addition, chemical reactions (between the oven, the crucible and uranium or uranium compounds) are more likely at higher temperatures. The following materials have been tested at LBNL using the 6.4GHz LBNL ECR (see Table 3): pure U, URe_2 , and UO_2 using W, Re or Ta oven crucibles with or without yttria ceramic liners. To achieve sufficient flux of uranium vapor pressures between $1 \cdot 10^{-4}$ mbar to $1 \cdot 10^{-2}$ mbar are required in the oven crucible. As reference, the vapor pressures for uranium and uranium dioxide are given in Table 2 as deduced from the literature [15-17]. U has a vapor pressure of about 10^{-2} mbar at 2200 C. UO_2 has a vapor pressure of about 10^{-2} mbar at 2000°C. The exact vapor pressure for URe compounds is not known, but experimentally the required temperature for URe_2 were found to be typically between 1750 (high charge states) to 2000°C (medium charge states).

Table.2 Vapor pressure for uranium and uranium dioxide^[15-17]

T [°C]	U	UO2	UO2
1500	$1.57 \cdot 10^{-6}$	$1.06 \cdot 10^{-6}$	$8.00 \cdot 10^{-6}$
1700	$4.50 \cdot 10^{-5}$	$6.43 \cdot 10^{-5}$	$5.36 \cdot 10^{-5}$
1900	$6.67 \cdot 10^{-4}$	$2.00 \cdot 10^{-3}$	$5.65 \cdot 10^{-3}$
2100	$6.09 \cdot 10^{-3}$	$3.66 \cdot 10^{-2}$	$6.56 \cdot 10^{-2}$
2300	$3.84 \cdot 10^{-2}$	$4.44 \cdot 10^{-1}$	$5.2 \cdot 10^{-1}$

As a result of the tests conducted at LBNL UO_2 was determined to be the most promising compound. The following paragraphs describe that choice based on the material tests conducted with various compounds and oven crucibles combinations (see also Table 3 and Table 4).

Table.3 Uranium beam production tests in the LBNL ECR ion source at 6.4 GHz

Oven crucible	W	Re	Ta	W + yttria	W + yttria
Uranium compound	UO_2	UO_2	URe_2	U	URe_2
Ion source	ECR, 6.4 GHz	ECR, 6.4 GHz	ECR, 6.4 GHz	ECR, 6.4 GHz	ECR, 6.4 GHz
Intensity (peak)	23 μA	11 μA	5-11 μA	8 μA	18 μA
Intensity (average)	12.5 μA	8 μA	Short term tests	Short term test	Short term tests
Charge state optimized	U^{18+}	U^{21+}	$\text{U}^{17+-21+}$	U^{24+}	U^{16+}
Hours	163 h (1 oven)	11 h	Short tests	12 h	12 h
Oven condition after	undamaged	Undamaged, used	Undamaged, used	Crystallization on	
Temperature	2100 C	1900-2000 C	1800-2000 C	1900 C	2150 C

Table 4. Uranium beam production tests in the VENUS ECR ion source using double frequency heating at 18 + 28 GHz

Oven crucible	Ta	Ta	Re
Uranium compound	URe_2	URe_2	UO_2
Intensity (peak)	205 μA	2-5 μA	3 μA
Intensity (average)	Short term tests		Cyclotron beam development
Charge state optimized	U^{33-34+}	U^{46-50+}	U^{47+}
hours	Short term test (<10h)	10 h	6 h
Oven condition after	Oven stems damaged	undamaged	Undamaged, test ongoing
Temperature	1900 - 2000 C	1750 C	1600 C

URe_2 was tested first, since it has been successfully used for uranium beams at LBNL before^[18] and has the lowest temperature requirements. As expected it was also successfully used in the LBNL ECR (Table 3) and in the VENUS ECR ion source (see Table 4). The highest intensity charge state spectrum achieved during these tests is shown in Figure 2. About 200 μA of U^{33+} and U^{34+} , and more than 175 μA of U^{35+} were extracted from the VENUS ECR ion source. However, one of the challenges with URe_2 is that it is not commercially available, and therefore we have been searching for alternative options.

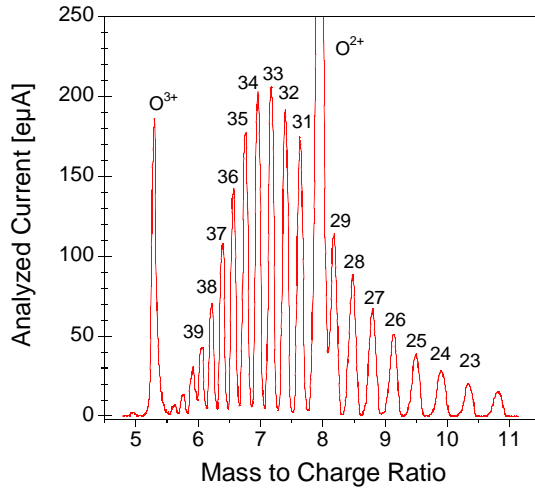


Fig. 2 Uranium CSD distribution for a high intensity medium charge state tune optimized for 33 to 34+

Pure uranium melts at 1135°C, but can be contained in pure Ytria crucibles and tungsten ovens^[19]. Its relatively low vapor pressure requires operating the oven at fairly high temperatures, at which yttria starts to sublime. Nevertheless, we were able to produce a low intensity beam using pure uranium in yttria crucibles for several hours, but pure uranium has been rejected as possible choice for high intensity beams.

UO₂ has a vapor pressure of about $1 \cdot 10^{-2}$ mbar at 2000°C^[17,20], therefore requiring temperatures between 1900 and 2100°C for the VENUS oven. If the oven can sustain these high temperatures for extended periods of time, it is the ideal compound for several reasons. It is chemically stable and easy to handle. UO₂ sublimates and has a very high melting point of 2820°C^[19]. In addition, its oxygen content is an ideal mixing gas for the plasma. To test the suitability of UO₂ as compound for producing uranium beams, long term tests were performed at the 6.4 GHz LBNL ECR (see Table 3). A tungsten oven was loaded with 530mg of UO₂ and operated for 96 hours until the oven was empty. The oven was reloaded with 317 mg of UO₂ and operated for additional 67 hours after which it was removed without visible damage from the ECR ion source. After this promising test series at the LBNL ECR, UO₂ tests are currently in progress with VENUS.

However, during the initial uranium tests in the VENUS ECR ion source the resistively heated oven bent and failed after a few hours of operation due to the presence of a strong $I \times B$ force in the high axial magnetic field of the VENUS injection solenoid.

While off-line oven performance tests and online uranium production tests at the LBNL 6.4 GHz ECR (Table 3) have shown that the oven assembly can reliably run at

furnace temperatures above 2200 °C for W, Re and Ta ovens, the oven experiences a strong $I \times B$ force when it is placed into VENUS. The axial magnetic field can be up to 4T at the oven and the magnetic field direction is perpendicular to the oven's current flow (see Fig 3 and Fig 4) [21].

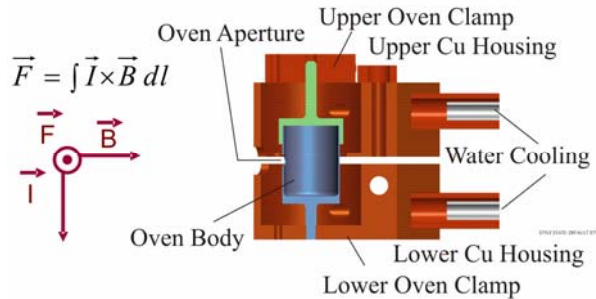


Fig.3 CAD representation of oven inside housing. A Lorentz force acts upon the unsupported lengths of the oven and can lead to bending of the oven leads.

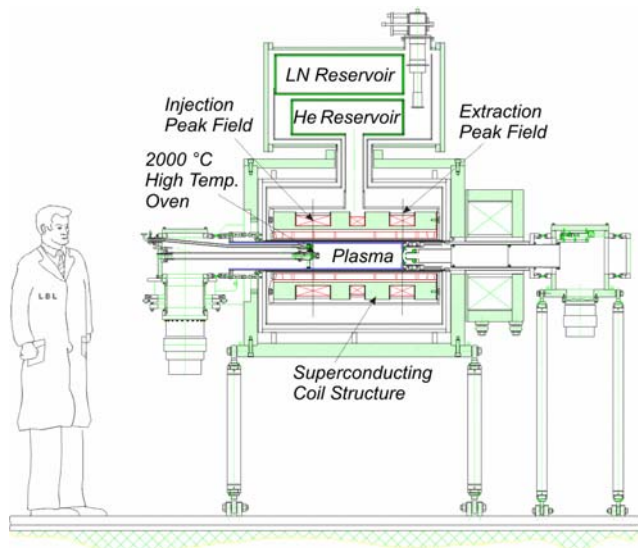


Fig.4 Mechanical layout of the VENUS ECR ion source. The location of the high temperature oven and the locations of the axial magnetic peak fields at injection and extraction of the ECR ion source are indicated.

At magnetic field strengths approaching 4T, and an oven current of 300A, the Lorentz force can reach 30N over the oven's unsupported axis. In addition, as the operational temperature and run time increase, the oven material's creep strength decreases [22]. To prevent bending of the hot oven crucible under the magnetic force leading to an oven failure, a coaxial oven design is presently in development, for which

the heater current flow will be parallel to the magnetic field.

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5. References

- 1 D. Hitz, A. Girard, G. Melin, S. Gammino, G. Ciavola et al., Rev. Sci. Instrum **73** (2), 509-512, (2002).
- 2 S. Gammino, G. Ciavola, L. Celona, P. Romano, L. Torrisi et al., Proceedings of the Cyclotrons and their Applications 2001, East Lansing, Michigan, AIP, 600, 2001 223-227
- 3 D. Leitner, C. M. Lyneis, T. Loew, O. Tarvainen, D. S. Todd et al., RSI **77** (03A302), 03A303-301, (2006).
- 4 C. M. Lyneis, D. Leitner, O. Tarvainen, D. Todd, S. Virostek et al., RSI **77** (03A342), (2006).
- 5 D. Leitner, C.M. Lyneis, S.R. Abbott, D. Collins, R.D. Dwinell et al., NIMB **235**, 486-493, (2005).
- 6 P. A. Zavodszky, B. Arend, D. Cole, J. DeKamp, G. Machicoane et al., Rev. Sci. Instrum. **77**, 03A334, (2006).
- 7 T. Nakagawa, Y. Higurashi, M. Kidera, T. Aihara, M. Kase et al., Rev. Sci. Instrum. **77**, 03A304, (2006).
- 8 G. Ciavola, S. Gammino, L. Celona, L. Torrisi, S. Passarello et al., RSI **77**, (2006).
- 9 D. Leitner, C.M. Lyneis, H. Koivisto, T. Ropponen, J. Ropponen et al., RSI (included in these proceedings), (2008).
- 10 M.A. McMahan, Nucl Instr Meth B **241**, 409, (2005).
- 11 National Research Council, http://www7.nationalacademies.org/bpa/RISAC_PREPUB.pdf, 2006.
- 12 D. Leitner, D.S. Todd, M.L. Galloway, and C.M. Lyneis, Journal of HEP&NP, Chinese Physical Society **31**, 1-7, <http://www.impcas.ac.cn/usr/ecr/web/index.files/proceedings/D.Leitner.pdf>, (2006).
- 13 H. W. Zhao, L. T. Sun, X. H. Guo, X. Z. Zhang, Z. M. Zhang et al., Journal of HEP&NP, Chinese Physical Society **31**, 8-12, <http://www.impcas.ac.cn/usr/ecr/web/index.files/proceedings/H.W.Zhao.pdf>, (2006).
- 14 D. Hitz, A. Girard, K. Serebrennikov, G. Melin, D. Cormier et al., RSI **75** (5), 1403, (2004).
- 15 *Handbook of Chemistry and Physics*, 75 ed. (CRC, 1994).
- 16 K. Bernhardt and K. Wiesemann, Plasma Physics **24** (8), 867 - 884, (1982).
- 17 G. T. Reedy and M. G. Chasanov, J. Nucl. Mater. **42** (3), 241-357, (1972).
- 18 Z.Q. Xie, Rev. Sci. Instrum **69** (2), 625, (1998).
- 19 C. Lau, EURISOL target and ion-source working group, http://www.ganil.fr/eurisol/targetgroupmeetings/legnaro_11-12-00/lau_u_th.pdf, (2000).
- 20 R. J. Ackermann, P. W. Gilles, and R.J. Thorn, J.Chem.Phys **25**, 1098-1097, (1959).
- 21 T. Loew, S.R. Abbott, M.L. Galloway, D. Leitner, and C.M. Lyneis, Proceedings of the PAC'07, Albuquerque, NM, JACoW,
- 22 M. Rieth, Forschungs-zentrum Karlsruhe, Institut für Materialforschung, paper 1951, <http://www.extremat.org>, (2005).

