CHILI – APPROACHING THE FINAL FRONTIERS IN LATERAL RESOLUTION AND SENSITIVITY – A PROGRESS REPORT.

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Introduction: With the discovery of isotopic anomalies in small samples like presolar grains, IDPs, or Stardust samples, or in even smaller sample volumes within such grains, the need for analytical techniques for isotope analysis in very small volumes became obvious. However, a sheer increase in lateral resolution is not sufficient, since the number of atoms of a given species and the ability to detect them within a small volume rapidly become limiting factors.

In SIMS (secondary ion mass spectrometry), the highest lateral resolution has been achieved with the NanoSIMS [1] using a Cs^+ primary ion beam with ~50 nm beam diameter or an O^- beam with ~200 nm beam diameter. With state-of-the-art gallium liquid metal ion guns, beam diameters of 2.5 nm have been realized. However, ionization efficiencies with such guns, which are mainly used as cutting tools in FIB (focused ion beam) instruments, are rather limited. And even when reactive primary ion species like Cs^+ or O^- are used, the useful yield – the ratio of detected to consumed atoms – only rarely exceeds 1 %. This mainly results from limitations in ionization efficiency during sputtering and is further reduced by limited mass spectrometer transmission and detection efficiency.

RIMS (resonance ionization mass spectrometry) uses the >99 % of the secondary particles that are not directly ionized during ion sputtering or laser desorption but are released as neutral atoms. After removal from the sample surface, they are efficiently and discriminantly ionized with lasers tuned to electronic resonances.

CHILI (Chicago Instrument for Laser Ionization), a new RIMS instrument, is presently under construction at the University of Chicago [2–4]. It is designed to significantly surpass its predecessors CHARISMA [5] and SARISA [6] with regard to both lateral resolution and sensitivity.

Since most features of CHILI have been described previously [2–4], this abstract will focus on new developments that took place over the last year.

Ion Optics: In order to achieve a targeted useful yield of 40–50 %, 3D simulations using ion optics simulation software (SIMION from Scientific Instruments Services, Inc.) showed that a transmission of >99.99 % can be expected for the CHILI time-of-flight mass spectrometer at a mass resolution $m/\Delta m$ of >1280 at FWHM (full-width half-maximum) and, more important, >450 at 10 % and >260 at 1 % peak heights, respectively. Rather long peak tails that showed up in previous simulations were sufficiently suppressed. Figure 1 shows a simulated mass peak for $^{98}Mo^+$. The mass resolution is more than sufficient, since selectivity in RIMS is achieved through resonance ionization. The remaining limitation for the useful yield results from the ~60 % active surface area of the microchannel plate detector.

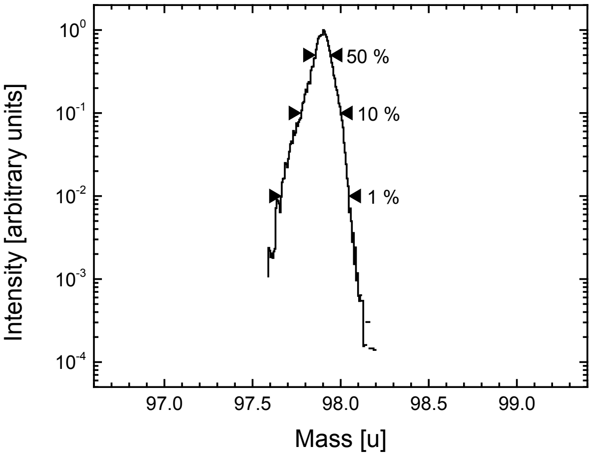

Fig. 1: *Simulated mass peak for 30,000 ^{98}Mo ions (ion package calculated according to [7]). Mass resolution $m/\Delta m$* is >1280, >450, and >260 *at 50 %, 10 %, and 1 % peak height, respectively.*

Ionization Lasers: Six tunable Ti:sapphire lasers for resonance ionization will be placed on the laser table as shown in Fig. 2. They will be pumped by three 40 W Nd:YLF lasers. This should allow routine isotopic analysis of at least three elements simultaneously. To increase the stability of the Ti:sapphire lasers, new holders for the Ti:sapphire crystals have been built that allow for very effective water cooling. In addition, any drift in wavelength of the tunable lasers is compensated by actively rotating the reflective diffraction grating in the laser cavities using piezoelectric actuators and feedback controls. This system has proven its excellent capabilities and is now routinely used with older instruments [8].

In order to use the laser photons more efficiently, all six beams will be made collinear with a three- or four-prism beam combiner and passed eight times through the vacuum chamber using multiple reflections from two Pellin-Broca [9] type prisms. This will effectively increase the size of the ionization volume by a factor of eight and allow the use of more focused beams (i.e., higher intensity) than in single path instruments, where the beam needs to be defocused to cover a large ionization volume.

Present Status: The present design status of CHILI is given in Fig. 2. The main vacuum chamber with its vertical flight tube is mounted on the center of an H-shaped laser table, 4.27 m × 3.66 m in size, which is supported by active piezoelectric vibration isolators. A Ga liquid metal ion gun (COBRA-FIB from Orsay Physics), which can be focused to 2.5 nm, is attached to the main chamber, as is a field-emission electron gun (e^-CLIPSE Plus also from Orsay Physics) and a secondary electron (SE) detector. The chamber is under vacuum ($<4\times10^{-9}$ hPa), and both guns and the SE detector are fully operational. The flight tube as well as most of the ion optics have been manufactured and await installation. A Schwarzschild optical microscope with a resolution of ~0.5 µm is almost ready for installation. The light optics will also be used to focus a UV desorption laser beam onto the sample. Three pump lasers are sitting on the table and most parts for Ti:sapphire lasers have arrived.

Summary: CHILI reflects many recent developments in instrument design, and most technical specifications are pushed towards their physical limits. After completion in the next few months, CHILI will be applied to a multitude of cosmochemical problems such as analysis of the most challenging samples from the Stardust mission.

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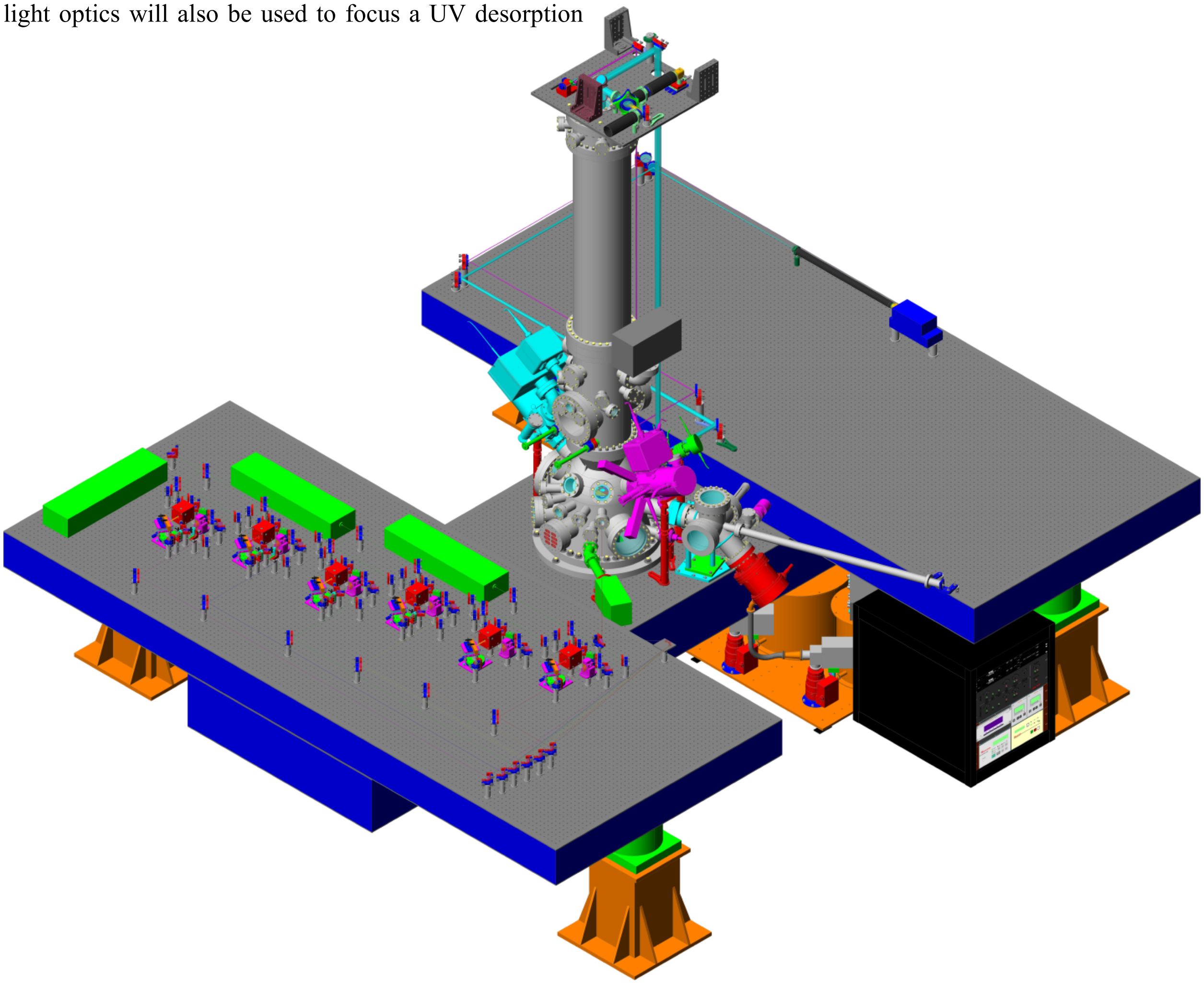

Fig. 2: *Present design status of the CHILI instrument as 3D CAD model.*