MAKING CHILI (CHICAGO INSTRUMENT FOR LASER IONIZATION)—A NEW TOOL FOR THE ANALYSIS OF STARDUST. T. Stephan[1,2,3], A. M. Davis[1,2,4], M. J. Pellin[1,2,3], M. R. Savina[2,3], I. V. Veryovkin[2,3], A. J. King[1,2], N. Liu[1,2,3], R. Trappitsch[1,2], and R. Yokochi[1,2], [1]Department of the Geophysical Sciences, University of Chicago, Chicago, IL 60637, USA, [2]Chicago Center for Cosmochemistry, [3]Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, USA, [4]Enrico Fermi Institute, University of Chicago, Chicago, IL 60637, USA. (tstephan@uchicago.edu)

Introduction: Recent sample return missions like Stardust [1] and Hayabusa, as well as presolar grains (real stardust) in meteorites and IDPs, have driven a strong desire for new analytical techniques with unprecedented lateral resolution and high sensitivity. Many of these samples are sub-micrometer in size and therefore just at the lower limit of present-day SIMS (Secondary Ion Mass Spectrometry) techniques [2].

The ultimate goal, to determine the isotopic nature and the location of every single atom in a given sample seems to be no longer beyond imagination. The final limiting factors are the number of atoms in an analyzed volume and how fast they can be detected.

Making CHILI (Chicago Instrument for Laser Ionization), a new RIMS (Resonance Ionization Mass Spectrometry) instrument, which is presently under construction at the University of Chicago, is a major step toward this goal [3, 4]. It builds on the success of two previous instruments of similar design, CHARISMA [5] and SARISA [6] but will surpass its predecessors with regard to lateral and mass resolution as well as sensitivity.

RIMS vs. SIMS: In SIMS, the useful yield—the ratio of detected to consumed atoms from a sample—only rarely exceeds 1 %, largely because of limitations in the ionization efficiency by sputtering. Limitations in mass spectrometer transmission and detection efficiency further reduce the useful yield. RIMS, however, makes use of the >99 % of the secondary particles that are not directly ionized in the sputtering process and which are released as neutral atoms. These atoms, after removal from the sample surface by ion sputtering or laser desorption, can be efficiently ionized with lasers tuned to electronic resonances.

TOF-MS: Efficient ionization, coupled with a high transmission time-of-flight mass spectrometer (TOF-MS), should lead to a useful yield of 40–50 % for CHILI, mainly limited by the ~60 % active surface area of the microchannel plate detector. Simulations using ion optics simulation software (SIMION from Scientific Instruments Services, Inc.) showed that a transmission of very close to 100 % can be expected for the CHILI TOF-MS. Further simulations using 3D models directly exported from the design and engineering software (AutoCAD from Autodesk, Inc.) are presently being performed to improve mass resolution of CHILI. Mass resolution for RIMS in general is less critical than for SIMS instruments, since selectivity is achieved through resonance ionization. However, for non-resonant ionization via single photons using a VUV F_2 excimer laser (157 nm), which is also envisioned for CHILI, mass resolution becomes important.

Major improvements in detection efficiency and mass resolution for CHILI compared to previous instruments will be realized by an increase of the acceleration voltage for the photoions from 1 kV to 9 kV.

Ion Gun: To achieve maximum lateral resolution, CHILI uses a new Ga liquid metal ion gun (COBRA-FIB) from Orsay Physics that can be focused to 2.5 nm, which, for >10 keV primary ions, is below the typical diameter of collision cascades achieved in ion sputtering. Further substantial increases in lateral resolution can therefore not be accomplished by ion sputtering.

Electron Gun: In addition to the ion gun, a new field-emission electron gun (e^-CLIPSE Plus) from Orsay Physics, which can be focused to 4 nm, is used for CHILI. Together with a UHV secondary electron detector also provided by Orsay Physics, CHILI operates as a fully functional field-emission scanning electron microscope for sample imaging.

Optical Microscope: A Schwarzschild optical microscope with a resolution of ~0.5 µm will also be implemented on CHILI. The light optical axis of the microscope is collinear with the ion optical axis for the secondary ions. This should assure higher mass resolution of the TOF-MS compared to previous instruments, where the secondary ion path was bent in order to separate the light path from the ion path.

In addition, the light path of the optical microscope can also be used to focus a UV desorption laser beam. Laser desorption can be applied to analytical problems where a lateral resolution of ~0.5 µm is sufficient. The UV laser can also deposit more energy in a spot than an ion beam, allowing the higher sample removal rates necessary for low abundance element detection.

Laser Ionization: For resonant ionization, tunable Ti:sapphire lasers will be used in CHILI. It is planned to initially have six such lasers, pumped by three 40 W Nd:YLF lasers. This should allow routine isotopic analysis of at least three elements simultaneously.

Ion Counting: Counting statistics limits the dynamic range in most current instruments. Most detectors cannot accurately count more than one ion per time channel for one ionization pulse, and count rates are typically limited to 0.1 counts per pulse for the most abundant ion species to avoid major dead time corrections [7]. For CHILI, a microchannel plate detector that will be able to distinguish multiple ions per pulse is presently under development, in collaboration with the Large-Area Picosecond Photo-Detectors Project (http://psec.uchicago.edu). If successful, this will increase the dynamic range by 2–3 orders of magnitude and decrease statistical errors by at least one order of magnitude for analyses that are not atom-limited.

Mechanical Design: In order to minimize relative motion of the various parts of CHILI, great emphasis was put on creating a rigid structure that is isolated as a whole from possible sources of vibration. The sample sits horizontally on a piezoelectric driven stage that provides sub-micrometer reproducibility. The sample holder holds samples up to ~45×65 mm^2 in size.

Vacuum System: In order to minimize noise and vibration, a vacuum system was designed that relies on magnetically levitated turbomolecular pumps with special vibration isolation. Analysis and sample transfer chambers are pumped independently. Each turbomolecular pump is backed by a drag pump that withstands relatively high fore-vacuum pressures up to 18 hPa. The drag pumps are backed by reservoir volumes that only need to be evacuated once per month. Even when major power failures occur, the vacuum chambers are completely isolated from all form of lubricants and hermetically sealed. Ultra-high vacuum (UHV) conditions (<10^{-9} hPa) will be easily achieved in the analysis chamber by using internal bakeout lamps.

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

References: [1] Brownlee D. et al. (2006) *Science, 314,* 1711–1716. [2] Stadermann F. J. et al. (1999) *LPS XXX,* #1407. [3] Davis A. M. et al. (2009) *LPS XL,* #2472. [4] Stephan T. et al. (2010) *LPS XLI,* #2321. [5] Ma Z. et al. (1995) *Rev. Sci. Instrum., 66,* 3168–3176. [6] Veryovkin I. V. et al. (2004) *Nucl. Instr. and Meth. B, 219–220,* 473–479. [7] Stephan T. et al. (1994) *J. Vac. Sci. Technol. A, 12,* 405–410.

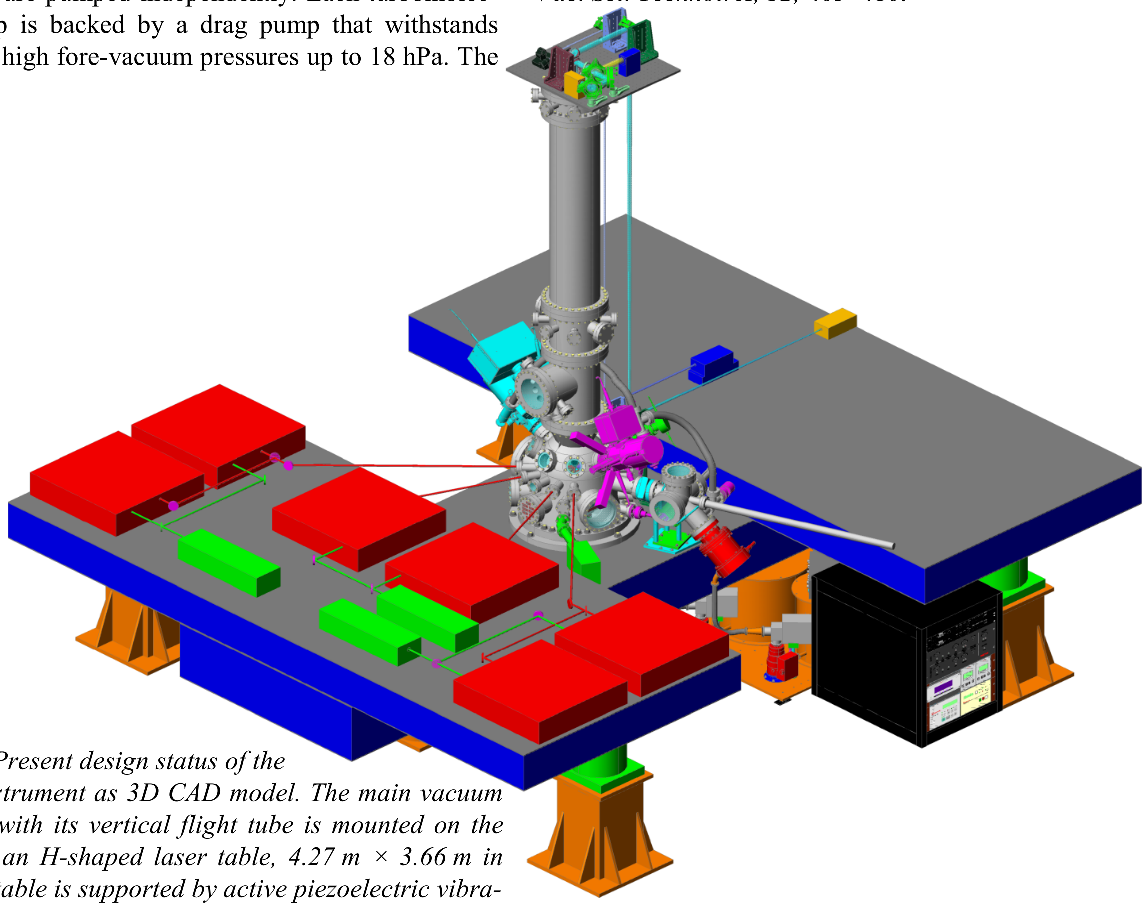

Figure: *Present design status of the CHILI instrument as 3D CAD model. The main vacuum chamber with its vertical flight tube is mounted on the center of an H-shaped laser table, 4.27 m × 3.66 m in size. The table is supported by active piezoelectric vibration isolators (green). The ion gun (magenta) and electron gun (cyan) lie on either side of the main chamber. Six Ti:sapphire lasers (red) and three pump lasers (green) fill one side (bottom left) of the laser table. On top of the ~2 m flight tube sits an optical breadboard with optical elements used for the Schwarzschild microscope. It allows sample observation and illumination as well as laser desorption. The camera (orange) and desorption laser (blue) are sitting on the other side of the laser table (top right), where there is also room for further expansion.*