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Isotope and Element Ratio Measurement with Femtosecond Laser Ablation Resonance Ionization Mass Spectrometry

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The Chicago Instrument for Laser Ionization (CHILI), a microbeam resonance ionization mass spectrometer, was originally built to desorb atoms from surfaces with a Ga^+ ion beam or a 20 ns pulse-length, 351 nm laser [1]. This configuration has been highly successful for isotopic analysis of µm-sized presolar SiC grains [e.g., 2,3] and selected other materials [4,5]. However, Ga^+ sputtering rates are limited by the number of Ga^+ ions that can be bunched into a well-focused 100 ns ion pulse. Therefore, laser ablation with its higher desorption rates becomes the method of choice for trace element analysis. With the long pulse-length laser, volatilization of neutral atoms occurs by surface heating. Samples must have strong 351 nm absorption and thermal effects must not cause local diffusion or surface modification. Even on materials that couple well, small changes in laser pulse energy lead to large changes in signal, because vapor pressure is exponential in temperature. Fluctuating ion signals hamper high precision, since dead time correction relies on constant count rates. To overcome these difficulties and expand the range of materials that can be analyzed, we installed a 190 fs pulse-length laser, capable of operating at 343, 515, or 1030 nm with excellent pulse-to-pulse stability. Only the shortest wavelength has been tested so far, but performance is excellent. We have measured element ratios (Ru/Mo) and isotopic compositions (Mo, Ru, and Ba) in a variety of materials, including some that are transparent to 343 nm light (silicate glasses and sapphire [6,7]). This opens CHILI to a wide variety of samples, including samples of asteroids and cometary dust returned by spacecraft, and meteorites and their constituents (e.g., presolar grains, refractory inclusions, chondrules, and fine-grained matrix).

[1] Stephan et al. (2016) *Int. J. Mass Spectrom.* 407, 1–15. [2] Stephan et al. (2019) *Astrophys. J.* 877, #101. [3] Liu et al. (2022) *Eur. Phys. J. A* 58, #216.[4] Trappitsch et al. (2018) *Astrophys. J.* 857, #L15. [5] Boehnke et al. (2018) *Proc. Nat. Acad. Sci.* 115, 6353–6356. [6] Regula et al. (2023) *Lunar Planet. Sci.* 54, #2988. [7] Korsmeyer et al. (2023) *Meteorit. Planet. Sci.* 58, #6256.