**A NEW IMAGING RESONANCE IONIZATION MASS SPECTROMETER FOR ISOTOPIC AND TRACE ANALYSIS***; M. J. Pellin[1], R. N. Thompson[1,2], Z. Ma[1], A. M. Davis[3], R. S. Lewis[3], and R. N. Clayton[2,3,4], [1]Materials Science/Chemistry Divisions, Argonne National Laboratory, Argonne, IL 60439, [2]Department of Geophysical Sciences, [3]Enrico Fermi Institute, [4]Department of Chemistry, University of Chicago, Chicago, IL, 60637

Chemical and isotopic analysis of meteorites, interplanetary dust particles, and other planetary materials increasingly requires microscopic determinations with an accuracy and precision not addressable with current instrumentation. The need for such measurements is clearly at odds with a desire to examine samples whose spatial extent is measured in nanometers. At Argonne National Laboratory, we have developed a resonance ionization mass spectrometer capable of both high useful yields ($>10^{-2}$) and trace analysis (<100 ppt). The instrument incorporates a high resolution optical microscope that can be used for both sample viewing (illumination is normal to the sample surface) and for laser ablation. There are a number of outstanding problems in the study of stardust that must await improvements in analytical technology: (1) isotopic analysis of many elements in SiC, including Zr, Mo, REE, in order to improve constraints on processes of nucleosynthesis; (2) isotopic analysis of Si and C in individual SiC grains <0.5μm across in order to see whether these grains sample populations not seen so far; (3) Si and Ti isotopic analysis of graphite at high precision, to learn more about the formation conditions of TiC inclusions in graphite; (4) improvement in data on trace element abundances and isotopic compositions of diamond; (5) more thorough searches for and characterization of stardust in situ in meteorites and in interplanetary dust particles collected from the stratosphere, Antarctica and Greenland. Some projects we hope to begin shortly while others will require significant development work before they can be attempted.

Our work has built on the extensive experience and equipment for Resonance Ionization Mass Spectrometry (RIMS) available at ANL. The new machine, named CHARISMA (CHicago Argonne Resonance Ionization Spectrometer for Micro-Analysis), has been constructed to overcome several problems in a previous machine, SARISA IV, which made isotopic analysis of meteoritic samples difficult. These problems include: (1) the difficulty of installing a high-quality microscope in SARISA IV, (2) problems with installation of small spot ion sources in SARISA IV; and (3) the desirability of a higher mass resolution time-of-flight spectrometer.

Historically, the most spectacular example of the power of microbeam analytical methods has been the revelation of details of nucleosynthetic processes from determination of the isotopic compositions of individual grains of circumstellar dust recovered from meteorites[1]. Diamond, silicon carbide and graphite that formed around stars and survived solar system formation and incorporation into meteorite parent bodies have been separated from carbonaceous chondrite meteorites and analyzed by a variety of techniques. They have been found to have anomalous isotopic compositions for every element analyzed in them. Different types of grains have isotopic signatures of different circumstellar environments. Analyses of individual grains reveal correlations between isotopic properties that reveal a great deal about nucleosynthetic processes. Stardust is extremely fine-grained. Diamond has an average grain size of only 16μm and each grain contains only 60 to 1100 atoms. Analyses of individual grains have not been performed because the grains are so small. Silicon carbide grains range from <0.05 to 20 μm across and individual grains greater than 0.5 μm across have been analyzed isotopically by ion microprobe for a limited suite of elements. Graphite grains are 0.8 to 10 μm across and occasionally contain TiC inclusions. Isotopic analyses of C, N, O, Si and Mg have been made by ion microprobe, but precisions of isotopic compositions of minor elements in graphite (O, Si, Mg) have been poor because of the limited sensitivity of the ion microprobe.

As a first step, we plan to measure titanium in individual presolar SiC grains separated from the Murchison meteorite. The titanium isotopic composition has been measured in some of these grains by conventional ion probe techniques[1]. The bulk of the grains have enrichments or depletions of all other isotopes relative to $^{48}$Ti. The magnitude of the effects, –40 to +160‰ in $\partial^{46}$Ti, –40 to +60‰ in $\partial^{47}$Ti, –40 to +220‰ in $\partial^{49}$Ti and –60 to +400‰ in $\partial^{50}$Ti (all normalized

to $^{48}$Ti) are adequate to be readily measured by the current generation RIMS-TOF. (Repetition of these measurements is essential to gain confidence in the new technique.) The isotopic pattern for titanium is roughly consistent with the expectations for a contribution from the He-shell *s*-process region of AGB stars. In detail, though, it cannot be that simple as the enrichments, and even the relative enrichments, of each of the isotopes are not constant. Instead, each grain has its own pattern. In addition, most of the data are not consistent in precise detail with the current theoretical production rates in AGB stars. These variations tell us something about the composition and nucleosynthetic history of the stars sampled by these grains, as well as the limitations of the current theory of stellar nucleosynthesis. It seems clear to us that we will have to make correlated measurements on several elements in each grain, if we hope to make progress in disentangling these clues. The higher efficiency of the new RIMS-TOF instrument compared to conventional ion probes will be essential in this endeavor.

**References:** [1] E. Anders and E. Zinner (1993) *Meteoritics* **28**, 490

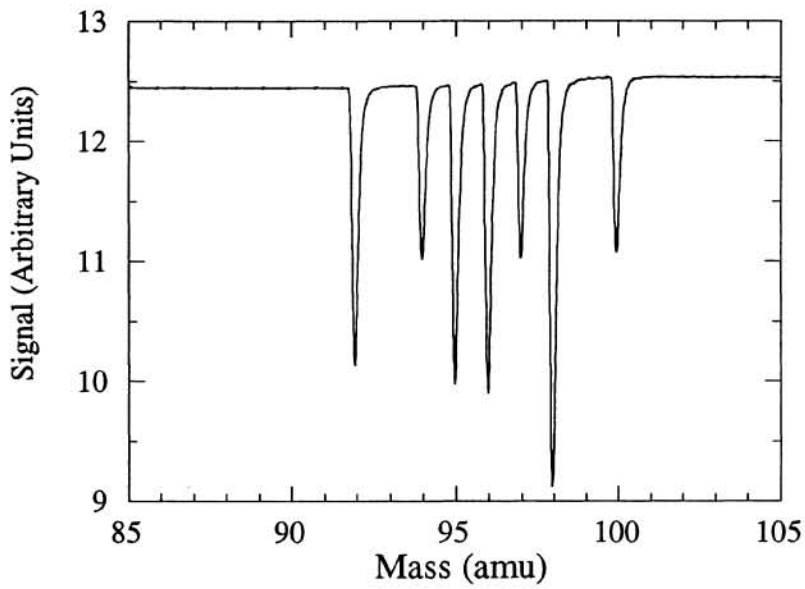


Figure (1): A preliminary mass spectrum obtained from laser ablation of a molybdenum sample. Mass resolution (m/Δm) is 400.

*Work supported jointly by the US. Department of Energy, BES-Materials Sciences, under Contract W-31-109-ENG-38 and by the Planetary and Materials Division of NASA.