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Presolar graphite grains and their stellar origins: A review

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Presolar grains are sub-micron to micron-sized stardust grains that are found in very primitive solar system materials. Laboratory measurements of presolar grains allow cosmochemists to understand the chemical and physical conditions in circumstellar environments, where the grains condensed. They provide important constraints for models of stellar nucleosynthesis and Galactic chemical evolution. In this paper, we review our current knowledge of the stellar sources of presolar graphite grains. This presolar grain species is not as abundant or widely studied as silicon carbide grains but is already known to have a variety of interesting stellar sources. Isotopic and microstructural measurements of more than 2000 graphite grains indicate that ~ 26% originate in Type II supernovae, ~ 48% in low-metallicity asymptotic giant branch (AGB) stars, and less than 1% in post-AGB stars and J-type stars. The remaining 25% of the grains have an ambiguous origin. We discuss the available evidence for the various stellar sources and outline the problems that are encountered while comparing grain data with astrophysical models.

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1. Introduction

Presolar or stardust grains are circumstellar condensates from the cooling outflows of dying stars. Sub-micron- to micron-sized stardust grains survived their journey through the interstellar medium, the collapse of the parent molecular cloud of the Sun, the accretionary processes of the early solar system and were finally incorporated into parent bodies of meteorites. Presolar grains are found embedded in the fine-grained matrices of the least thermally altered chondrites, interplanetary dust particles (IDPs), and Antarctic micrometeorites (AMMs). These grains can be identified on the basis of their exotic isotopic compositions that are representative of those of the atmospheres of the parent stars, at a given time in their evolution. The isolation of presolar grains in 1987 marked the beginning of a very important subfield of laboratory astrophysics. Isotopic, elemental, and structural measurements of presolar grains provide us with an unparalleled opportunity to study the chemical and isotopic compositions of individual stars at a level of precision that cannot be attained by astronomical observations. Studying these grains provides important constraints to stellar nucleosynthesis and evolution models. The grains also contain vital clues to the chemical and physical properties of stellar atmospheres, Galactic chemical evolution, interstellar and nebular processes, and early solar system conditions and processes.

2. Types of presolar grains

A "burn the haystack to find the needle" approach was used to separate the first presolar grains. This consisted of a series of harsh acid dissolution steps to isolate the acid-resistant carrier phases of extreme noble gas anomalies. Presolar diamonds were first separated by isolating the carrier of the noble gas component, Xe-HL [1]. Subsequently, silicon carbide grains were found to be the carriers of Ne-E(H) and Xe-S [2,3] and graphite, the carrier of Ne-E(L) [4,5]. Other acid resistant presolar phases, that do not carry large noble gas anomalies, are oxides (spinel ($MgAl_2O_4$), corundum (Al_2O_3), and hibonite ($CaAl_{12}O_{19}$)) [6,7]. Presolar silicates were discovered by carrying out automated, *in situ* raster ion imaging of meteorites and IDPs [8−11]. Additionally, silicon nitride grains [12] and subgrains within presolar grains (Ti, V, Mo, Zr, Ru carbides, kamacite and Fe grains, sulphides, silicides etc.) are the other presolar grain phases that have been identified to date. A recent, detailed review of the different presolar grain types can be found in [13] and references therein.

In this paper, we focus on presolar graphite grains and discuss their stellar sources.

3. Presolar graphite grains

Presolar graphite grains were first isolated by Amari et al. [4,5] from the Murchison CM2 meteorite as the carrier phase of Ne-E(L) (almost pure ^{22}Ne). Subsequently, extensive multi-element isotopic studies have been carried out on 1850 graphite grains from Murchison [e.g., 14−18] and 357 grains from Orgueil [e.g., 19−21]. In both the meteorites, spherical, carbonaceous grains, ≥ 1 μm in size were found to be presolar [5,17,21]. The inferred abundance of graphites from noble gas measurements is 1-10 ppm in primitive meteorites [22].

The isotopic properties of the grains are density-dependent and are used to classify grains from both Murchison and Orgueil as low- and high-density (LD and HD) grains. Murchison LD fractions are KE3 (1.6−2.05 g cm^{-3}) and KFA1 (2.05−2.10 g cm^{-3}) and those of Orgueil are OR1c (1.67−1.75 g cm^{-3}) and OR1d (1.75−1.92 g cm^{-3}). HD fractions of Murchison are KFB1 (2.10−2.15 g cm^{-3}) and KFC1 (2.15−2.20 g cm^{-3}) and those of Orgueil are OR1f (2.02−2.04 g cm^{-3}), OR1g (2.04−2.12 g cm^{-3}), and OR1i (2.16−2.30 g cm^{-3}). Note that there is not much overlap in the actual densities of the various fractions of the two meteorites but the isotopic properties of grains from LD and HD fractions from both the meteorites (discussed below) are very similar.

3.1 Isotopic properties and the stellar sources of presolar graphite grains

3.1.1 LD grains from Type II supernovae

Forty-six percent of grains from Murchison's LD fractions (KE3 and KFA1) [15] and 39% of LD grains from Orgueil (OR1c and OR1d) have isotopic signatures that indicate an origin in core collapse (Type II) supernovae (SNe). Bulk noble gas analyses of Murchison graphite separates [14] and single grain studies [23,24] of He and Ne isotopes find that Ne in the grains is highly enriched in ^{22}Ne but ^{4}He is low. Amari [25] concluded that the ^{22}Ne in LD graphites of Murchison comes from the decay of the short-lived radionuclide ^{22}Na ($\tau_{1/2}$ ~ 2.6 a) produced in the O/Ne zone of a SN. The low ^{4}He/^{22}Ne ratios and ^{18}O and ^{28}Si excesses measured in individual LD grains support this argument.

Most LD grains from both meteorites have ^{18}O excesses that are often accompanied by ^{15}N excesses [20,26]. Partial He-burning and explosive nucleosynthesis make ^{18}O and ^{15}N in the He/C zone of a Type II SN, which is the only zone where these excesses occur together. Isotope imaging of microtomed slices of LD grains from Orgueil shows regions with large correlated ^{15}N and ^{18}O excesses [27] confirming that these excesses originate from the same zone. Other isotopic properties of LD grains are very similar to those of SiC-X grains that are known to have a core-collapse SN origin [28]. Some LD graphite grains have inferred ^{26}Al/^{27}Al ratios (from large ^{26}Mg excesses in the grains; $\tau_{1/2}$ - ^{26}Al ~ 7.3×10^{5} a) as high as

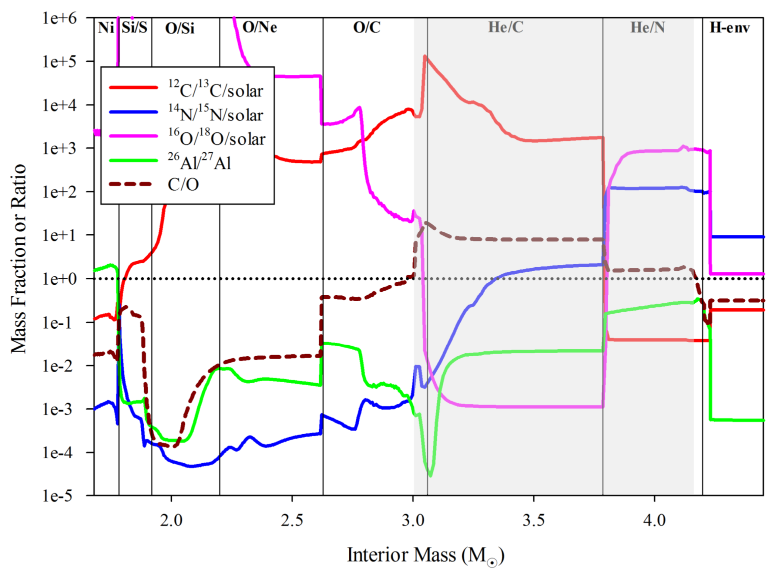

Fig. 1. Theoretical profiles of ^{26}Al/^{27}Al ratio and solar-normalized C, N, O isotopic composition in the interior of a 15 $M_{\odot}$ pre-explosion SN model [29]. The layer names are indicated along the top of the plot. The shaded region indicates layers where C > O. Mixing between the He/C and He/N zones cannot reproduce the isotopic compositions of some graphite grains as described in the text.

those observed in SiC-X grains ($^{26}Al/^{27}Al \sim 1$). Amongst the C-rich zones, the highest $^{26}Al/^{27}Al$ ratios are obtained in the He/N layer of a Type II SN (Fig. 1). A few grains show evidence for the presence of ^{41}Ca ($\tau_{1/2} \sim 1.05\times10^5$ a) inferred from ^{41}K excesses [30]. The $^{41}Ca/^{40}Ca$ ratios range from 0.001 to 0.01 and are higher than those predicted for AGB star envelopes [31]. Such high values are achieved in the C- and O-rich layers of Type II SNe. Further, some of the LD graphite grains have ^{28}Si excesses that are sometimes accompanied by large ^{44}Ca excesses. Large ^{44}Ca excesses (compared to the $^{40,42,43}Ca$ excesses) are attributed to the decay of ^{44}Ti ($\tau_{1/2}$ = 60 a) [32]. No other stellar source can explain correlated ^{28}Si excesses and the presence of ^{44}Ti. These isotopic signatures indicate contribution of material from the Si/S layer in the interior of a Type II SN.

In order to explain anomalies in individual graphite grains, several studies [e.g., 15,18,21,33] have performed mixing calculations between different SN. These calculations are justified by X-ray observational data of SN remnants [34] like Cassiopeia A that indicate extensive mixing in SN ejecta. As more isotopic systems are measured on individual graphite grains, such SN layer mixing exercises become extremely complex and more importantly, fail to reproduce all the isotopic anomalies observed in some grains. Mixing calculations for graphites and other carbonaceous grains require the largest contribution of materials to be from the C-rich layers of the SN to maintain a C/O ratio > 1 required for condensation. Theoretical models are unable to explain high $^{12}C/^{13}C$ and $^{26}Al/^{27}Al$ ratios in individual grains because these signatures arise from the ^{12}C-rich He/C zone, where $^{26}Al/^{27}Al$ is low, and the ^{13}C-rich He/N zone, where $^{26}Al/^{27}Al$ is high (Fig. 1). The low $^{12}C/^{13}C$ ratio measured in some grains does not allow mixing of too much material from the He/C (^{12}C-rich) zone to explain the large, measured ^{18}O and ^{15}N excesses in the grain (Fig. 1) [21]. Similar problems arise while explaining anomalies in SiC grains [35,36]. Jadhav et al. [21] discuss these problems in detail, as well as the limitations of various SN nucleosynthesis models and how they fall short of explaining grain data.

Pignatari et al. [37] recently offered an alternate solution to the mixing problem for ^{12}C-enriched, carbonaceous grains. According to their calculations, the shock wave passing through the layers after core-collapse causes explosive burning conditions to create a C- and Si-rich layer at the bottom of the He/C zone. This layer produces ^{28}Si by alpha capture reactions and, at higher energies, ^{44}Ti. At lower energies, however, the high $^{44}Ca/^{40}Ca$ ratios measured in grains probably arise due to large depletions in ^{40}Ca and the production of ^{44}Ca due to neutron capture reactions. A similar argument can be made for high $^{26}Mg/^{24}Mg$ ratios where the ^{26}Mg excesses measured in grains are from nucleosynthetic ^{26}Mg and depletion of ^{24}Mg, due to neutron capture reactions, and not the decay of radioactive ^{26}Al. Thus, if ^{18}O, ^{15}N, ^{26}Mg, ^{28}Si, and ^{44}Ca excesses are all created in the same explosive He/C-C/Si zone of SNe then contributions from other layers are not required. However, the problem with ^{13}C-enriched grains (described previously) still remains.

Additionally, transmission electron microscopy (TEM) studies of ultramicrotome sections of LD graphites reveal sub-grains of iron-metal phases and TiC that exhibit SN signatures [38]. One unique LD grain was found to contain all the phases that are predicted by equilibrium thermodynamic calculations for carbonaceous SN layers [39].

Thus, a majority of LD graphite grains originate in SNe. The number of SN grains decreases drastically in the HD fractions of both Murchison (2%) and Orgueil (7%) [15,21].

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There is no clear evidence for AGB grains among LD graphites from either Murchison or Orgueil.

3.1.2 HD grains from AGB stars

The HD grain fractions of graphites in Murchison and Orgueil are characterized by larger than solar $^{12}C/^{13}C$ ratios (up to 5000), terrestrial N, solar O isotopic ratios, and ^{30}Si excesses. The N and O isotopic compositions of HD graphite grains are puzzling in view of the fact that a majority of the same grains have $^{12}C/^{13}C$ ratios that vary over a wide range. The N and O isotopic compositions are therefore not considered to be intrinsic to the grains but the result of isotopic equilibration that took place either on the parent body or in the laboratory [15,17,19-21,40].

One of the *s*-process components of Kr, Kr-SH, with a high $^{86}Kr/^{82}Kr$ ratio resides in the HD KFC1 fraction of Murchison. It seems to originate in low-metallicity AGB stars, $Z \leq 0.002$ [14]. Kr-SH is not present in the KFB1 fraction. Combining Kr isotopic data with light noble gas studies [24,41] and C isotopic ratios, Amari et al. [42,15] concluded that KFB1 HD graphites from Murchison with large $^{12}C/^{13}C$ ratios ($\geq$ 100) originated in low-mass (1.5−3 $M_{\odot}$), low-metallicity (Z = 3−6×10^{-3}) AGB stars while KFC1 grains with $^{12}C/^{13}C \geq 60$ have the same sources as KFB1 graphites and also, 5 $M_{\odot}$ AGB stars with Z = 0.01 and 0.02. In the absence of Kr measurements for the Orgueil density separates, the only evidence for an AGB origin are high $^{12}C/^{13}C$ ratios that are sometimes correlated with large ^{30}Si excesses in a majority of the HD grains (Fig. 2). Such signatures are predicted for low-metallicity AGB stars where more ^{12}C and ^{30}Si are dredged up into the envelope, during the thermally pulsing (TP) phase, than in stars of solar metallicity (Fig. 2). Such stars have envelopes with high C/O ratios that might preferentially condense graphite over SiC, explaining the absence of SiC grains with high $^{12}C/^{13}C$ ratios and ^{30}Si excesses in the presolar grain inventory [19,44,45]. This argument is supported by astronomical observations by Leisenring et al. [46] and Sloan et al. [47] who find that the abundance of SiC grains in circumstellar dust decreases with decreasing stellar metallicity and is explained by the increasing C/O ratio. Calculations by Gail et al. [48] also predict that a large fraction of graphite grains condense around low-metallicity (~ 0.3 $Z_{\odot}$) AGB stars.

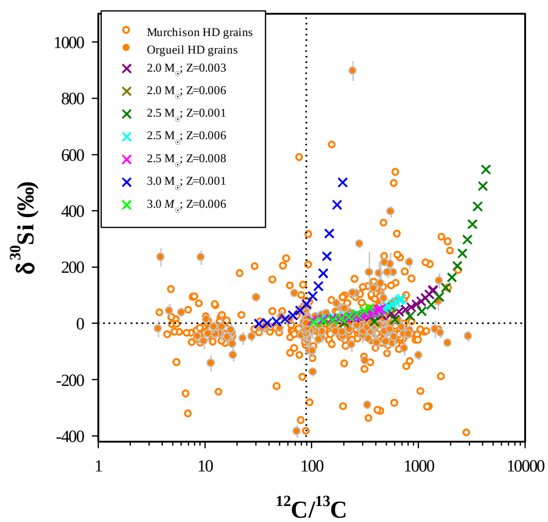

Fig. 2. C and Si isotopic compositions of HD graphite grains compared with F.R.U.I.T.Y. AGB models [43]. ^{12}C and ^{30}Si excesses indicate an origin in low-metallicity AGB stars. Dashed lines indicate solar values.

Further evidence for low-metallicity AGB stars as the stellar sources for HD graphite grains is provided by Zr, Mo, and Ba isotopic ratios in Murchison (KFC1) graphites [49] and OR1f Orgueil HD graphites [50,51] measured by resonant ionization mass spectrometry (RIMS) with the CHARISMA instrument. *S*-process signatures in Zr, Mo, Ba isotopes in some of the HD graphites indicate low-metallicity AGB origins for those grains. The new RIMS machine, CHILI, will greatly expand the very meagre, existing dataset of heavy-element isotopic data on graphite grains [52]. Co-ordinated light- and heavy-element isotopic analysis of presolar graphite grains will not only help determine the stellar sources of these grains but also provide important constraints to nucleosynthesis models (e.g., work by Liu et al., [53] constrains the rate of the ^{22}Ne(α,n)^{25}Mg reaction in AGB stars using Ba isotopes measured in mainstream SiC grains).

Furthermore, TEM studies [54] find HD grains to have internal subgrains of TiC that have high concentrations of the *s*-process elements Mo, Zr, and Ru that strengthen the argument for AGB origins of the parent grains. Croat et al. [55] used the composition of refractory metal nuggets (RMNs) found in HD graphites to derive a higher condensation temperature for HD graphites than for LD graphites.

Seventy-five percent of the Murchison HD grains are from AGB stars, mostly low-metallicity AGB stars, based on their He, Ne, Kr, C isotopic compositions [15]. In the absence of any noble gas measurements on individual HD grains or separate density fractions, we used the same classification criteria based on ^{12}C/^{13}C ratios as [15] and the presence of ^{30}Si excesses (along with AGB envelope values of ^{26}Al/^{27}Al and ^{41}Ca/^{40}Ca ratios in a few grains) and found that 64% of HD graphites from Orgueil likely originated in low-metallicity AGB stars.

3.1.3 HD grains from post-AGB stars

Among both Murchison and Orgueil graphite separates, a small population of ~ 10−15 % of the grains has ^{12}C/^{13}C < 20 [15,20,21,56]. Except for a few grains with unambiguous SN signatures, the stellar sources of these highly ^{13}C-enriched grains were puzzling for a long time. Amari et al. [57] suggested J stars or born-again AGB stars to be the sources of similarly ^{13}C-enriched SiC-AB grains. Jadhav et al. [20] found a few ^{13}C-enriched HD grains from Orgueil that have extremely large excesses in ^{42,43}Ca and $^{46, 47, 49, 50}$Ti that matched those predicted for the He-shell of AGB models. This study hypothesized that the grains probably originate from born-again AGB stars that suffer a very late thermal pulse (VLTP). A comparison of C, Ca, and Ti isotopic data of these HD graphite grains with VLTP nucleosynthesis calculations for Sakurai's object [58] strongly supported the born-again AGB star hypothesis and identified the first *i*-process signatures in presolar grains (Fig. 3) [56]. The low C isotopic ratios are a direct signature of H-ingestion, producing ^{13}C in the outer layers of the C-rich intershell in post-AGB stars. The Ca and Ti anomalies in 6 grains are, instead, due to the activation of the *i*-process at the bottom of the He intershell where neutron densities reach ~10^{15} cm^{-3} [58]. The ^{46,48}Ti isotopic data require that contributions of the irresolvable (by SIMS) isobars ^{46,48}Ca need to be taken into account to explain the anomalies in the grains. ^{46,48}Ti are destroyed during VLTP nucleosynthesis but abundant amounts of ^{46,48}Ca are produced during *i*-process nucleosynthesis in such stars. This claim can be further verified by measuring Ca isotopes by RIMS where isobaric interferences can be suppressed. Putative graphite grains from such stars should exhibit strong *s*-process and/or *i*-process signatures in elements such as Sr, Zr, Mo, Ru, and Ba that can be measured by RIMS.

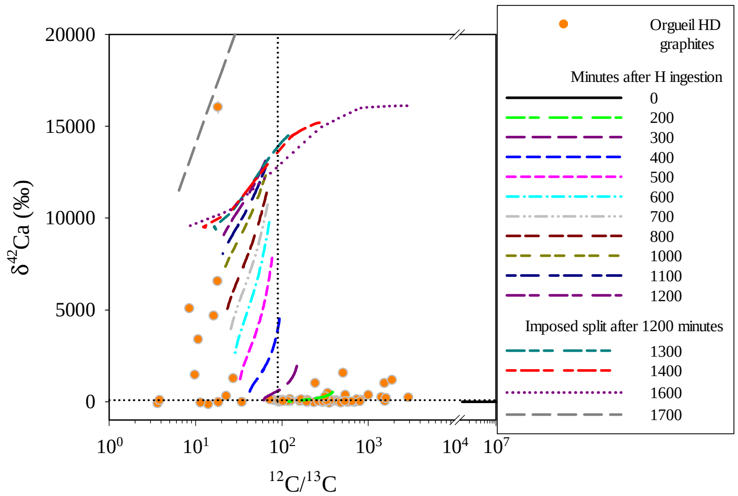

Fig. 3. Ca and C isotopic ratios measured in HD presolar graphite grains compared to VLTP nucleosynthesis model predictions [58]. Each line represents an isotopic depth profile calculated by the VLTP models of the He intershell at a given time (in minutes) after the start of H-ingestion. Grains with extreme Ca anomalies are ^{13}C-enriched while those with moderate anomalies are mostly ^{12}C-enriched. Dashed lines indicate solar values.

Jadhav et al. [56] also pointed out that the $^{12}C/^{13}C$ ratios in the VLTP models vary from 10 to 10^4. Thus, grains that display *i*-process signatures from post-AGB stars that suffered a VLTP do not have to be highly ^{13}C-enriched but can also be ^{12}C-enriched. The evidence for grains with VLTP signatures brings to light the fact that grains from post-AGB stars that suffer a late-thermal pulse (LTP) should also exist. In LTP post-AGB stars, material from the He intershell is brought up to the star's surface during the LTP but no ingestion of H into the intershell might occur. Such stars could have ^{12}C-enriched surfaces combined with He-intershell abundances of Ca and Ti isotopes (or, other neutron rich isotopes). These stars could be the sources of grains with moderate Ca and Ti anomalies and high $^{12}C/^{13}C$ ratios (Fig. 3) [56]. Two mainstream SiC grains were also found to exhibit *i*-process signatures in the Ba isotopes [53]. The Si isotopic compositions of these grains, however, do not agree with VLTP models. Post-AGB stars that underwent a VLTP have also been proposed as possible stellar source for three SiC AB grains that contain large ^{32}S excesses [59]. The VLTP models [58] predict production of radioactive ^{32}Si that decays to ^{32}S in such stars, making born-again AGB stars viable sources for some AB grains with ^{32}S excesses. Future measurements of heavy element isotopes by RIMS will determine the fraction of graphite and SiC grains that originated in VLTP and LTP post-AGB stars. A comparison of the abundance of carbonaceous grains from post-AGB sources that have suffered a VLTP or LTP and an estimate of the fraction of dust ejected by these stars will be the next step in future investigations by both the grain community and astronomers.

3.1.4 Other stellar sources

The first measurements of N isotopic ratios of Galactic J-type stars with $^{12}C/^{13}C$ ratios of 2−10 were carried out recently [60]. The study indicates that the $^{14}N/^{15}N$ ratios in these stars are both, much higher and lower than the terrestrial value of 272. This new result allows us to consider J-type stars as possible stellar sources of two ^{13}C-enriched HD graphite grains [21] with higher than terrestrial $^{14}N/^{15}N$ ratios whose sources have been ambiguous so far. In order to unambiguously assign J-type stellar sources to grains additional diagnostic isotopic signatures are required.

The recent study of superluminous or 'Super-Chandrasekhar" Type Ia SNe claims that the presence of strong C II lines in the spectra of two Type Ia SNe indicate that these objects were likely to form C dust [61]. Assuming that these stars do condense dust, the study estimates a mass range of ~ $(1-4)\times10^{-4}$ $M_{\odot}$, which is comparable to the mass condensed around Type II SNe. Future mid-IR observations of superluminous SNe Ia during the nebular phase will determine whether dust forms around these objects and if such SNe Ia can be considered to be sources of presolar dust. Several SN zone mixing problems encountered while explaining grain anomalies can be avoided if Type Ia SNe can condense carbonaceous dust [62 and references therein].

3.1.5 Summary of stellar sources

Of the nearly 2200 presolar graphites (from both Murchison and Orgueil) measured in the laboratory, ~26% appear to be SN condensates, and 48% probably originated from AGB stars (mostly low-metallicity AGB stars). Less than 1% of the grains show signatures of *i*-process nucleosynthesis from post-AGB stars and the remaining have an ambiguous origin. These numbers are expected to change once neutron-rich isotopes of heavy elements are measured with CHILI [52] in the near future.

4. Future perspectives

Multi-element isotopic studies of individual grains with SIMS play an important role in determining the stellar sources of presolar grains. Future multi-technique analyses on individual grains, already measured in an ionprobe, will help provide even tighter constraints on stellar chemical and physical conditions. Presolar graphite grains are ideal for multi-technique analyses because they are larger than other presolar grains species (~ average diameter = 3−4 μm). Such techniques include heavy-element isotopic measurements at very high-spatial resolutions (~ 10 nm) with the next-generation RIMS instrument, CHILI. This machine will be able to measure isotopic compositions of not just individual grains but also the subgrains within. See Davis et al. (this proceeding) and [52] for descriptions of CHILI's analytical capabilities and future plans for presolar grain work. High spatial resolution x-ray fluorescence and diffraction measurements with a synchrotron beam at ESRF and DESY will provide trace element information at the ppb level, as well as structural information on grains and their subgrains. This technique is powerful yet non-destructive, an important combination when attempting multi-technique measurements on micron-sized grains [63].

Exciting discoveries are expected in this relatively new field of laboratory astrophysics as innovative, cutting edge analytical techniques are developed or adapted for presolar grain research.

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