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# **Collaborators (past and present)**

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- Christian Vollmer Westfälische Wilhelms-Universität Münster (Münster, Germany)
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- Marco Pignatari University of Hull (U.K.)
- Wataru Fujiya Ibaraki University (Mito, Japan)



# Outline of the talk

- Introduction: What are presolar grains and how are they found?
- What can we learn by studying presolar grains?
- Tools of isotope analysis of presolar grains
- The parents of presolar grains: Stellar evolution and nucleosynthesis
- Presolar grain types: mineralogy, abundance, isotopy, structure
- What can we expect in presolar grain research in the near future?



Crab Nebula by Hubble © NASA, ESA, J. Hester

IRAS 05437+2502 by Hubble © ESA/Hubble, R. Sahai and NASA

TW Hydrae and associated disc by ALMA © S. Andrews (Harvard-Smithsonian CfA); B. Saxton (NRAO/AUI/NSF); ALMA (ESO/NAOJ/NRAO)

- Winds of red giants
- Stellar explosions

Interstellar medium (ISM)

Few nm to several µm diameter stellar dust grains that **pre**-date the Solar System, and partially or completely survived early Solar System processing: fossil, extra-Solar System stellar dust for direct study in the lab

600 nm Presolar SiC from the Murchison meteorite

Earth/Laboratory

Nascent Solar System

Primitive Solar System materials

A piece of the Murchison meteorite (Field Museum, Chicago, U.S.A.)

NanoSIMS at the Max Planck Institute for Chemistry (Mainz, Germany)



- Most grains are small (diameter <250 nm)</li>
- Silicates, oxides, graphite, SiC, etc.
- Found in meteorite matrices, interplanetary dust particles (IDPs) and cometary dust, based on *anomalous isotope composition* (i.e., composition outside the range of Solar System isotope variations) – sampling bias?
- First presolar isotope anomalies detected in bulk samples (H, O, *Ne, Xe*)
- First presolar grains detected in chemically separated aliquots of primitive meteorites





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Ernst Zinner\*, Tang Ming† & Edward Anders†‡



## Why presolar grains?

They document stellar evolution and Galactic Chemical Evolution

E.g., Si isotope model predictions for AGB stars...





## Why presolar grains?



They tell about dust formation in stellar environments...

- Pressure and T of condensing gas
- Condensation sequence(s)
- Kinetics of condensation

...and about ISM/early Solar System processes:

- Condensation of additional material
- Grain alteration/destruction



#### Polymineralic AGB condensate (Leitner et al., 2018)



AGB spinel aggregate with silicate mantle that formed in the ISM or the Solar System (Zega et al., 2020)

# Why presolar grains?



CC

CR2

CB3

CM2

CV3 CO3

CK4

1.5

Tringuier et al. (2007)



#### <sup>54</sup>Cr-enriched spinel grains

Mars

EC

0

E(54Cr)

0.5

1



## What kind of data?



- Isotope composition
- Chemical composition
- Structural data
- Relative abundance



FE-SE image of a presolar silicate (Leitner et al., 2018)

- Isotope/chemical/structural analysis of individual presolar grains requires *nano-analytical techniques:*
- Isotope compositions usually measured by nanoscale secondary ion mass spectrometry (the instrument is abbreviated NanoSIMS) and by resonance ionization mass spectrometry (RIMS; instruments: CHILI, LION, CHARISMA)
- Chemical composition determined by Auger spectroscopy and energy dispersive X-rax spectroscopy (EDX)
- Structural data obtained using transmission electron microscopy (TEM)

#### **Isotope** analysis



- I. NanoSIMS:
- Secondary ion mass spectrometer
- Two primary ion sources
  (Cs<sup>+</sup>, Hyperion: O<sup>+</sup> or O<sup>-</sup>)
- Spatial resolution down to 80 nm
- 5 masses analysed simultaneously
- Mass resolving power (MRP): several thousand (hydride interferences of major elements are easily resolved)
- Precision (isotope ratios of major elements in presolar grains): % level



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) = Presolar grain with isotope anomaly

#### **Isotope** analysis



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#### II. RIMS:

- Ionization of selected elements
- Ion or laser beam used to sample analyte
- Currently ~1 μm spatial resolution
- Low mass resolution (MRP <~1500), high useful yield (up to ~10 %)
- Some isotope systems are not accessible due to insufficient ionisation
- Isobaric interference problems
  circumvented
- Precision (isotope ratios of major elements in presolar grains): % level or better



CHILI, The University of Chicago

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|----|----|---|-----|-----|----|-----|---|------|----|-----|------|-----|------|--------|-----|----|-----|--|
| Li | Be | B C N O F M   |     |     |    |     |   |      |    |     |      |     |      |        |     | Ne |     |  |
| Na | Mg | be analysed with CHILI AI Si P S CI                 |     |     |    |     |   |      |    |     |      |     | Ar   |        |     |    |     |  |
| к  | Ca | Sc  | Ti  | V   | Cr | Mn  | Fe  | Co   | Ni | Cu  | Zn   | Ga  | Ge   | As     | Se  | Br | Kr  |  |
| Rb | Sr | Y   | Zr  | Nb  | Мо | Тс  | Ru  | Rh   | Pd | Ag  | Cd   | In  | Sn   | Sb     | Те  | Т  | Xe  |  |
| Cs | Ва | La  | Hf  | Та  | W  | Re  | Os  | Ir   | Pt | Au  | Hg   | TI  | Pb   | Bi     | Po  | At | Rn  |  |
| Fr | Ra | Ra Ac Elements key to detection of pre-solar grains |     |     |    |     |   |      |    |     |      |     |      |        |     |    | ins |  |
|    |    |   |     |     |    |     | AI    Si    P    S    CI    Ar      Mn    Fe    Co    Ni    Cu    Zn    Ga    Ge    As    Se    Br    Kr      Tc    Ru    Rh    Pd    Ag    Cd    In    Sn    Sb    Te    I    Xe      Re    Os    Ir    Pt    Au    Hg    TI    Pb    Bi    Po    At    Rn      Key to detection of pre-solar grains    Re    Stephan et al. (2016)    Stephan et al. (2016)    Ar |      |    |     |      |     |      |        |     |    |     |  |
|    |    |   | Се  | Pr  | Nd | Pm  | Sm  | Eu   | Gd | Tb  | Dy   | Ho  | Er   | Tm     | Yb  | Lu |     |  |
|    |    |   | Th  | Pa  | U  | Np  | Pu  | Am   |    |     | Sten | han | et o | ıl. (2 | 016 | )  |     |  |

## **Chemical analysis**



- SEM-EDX (Scanning Electron Microscope-Energy Dispersive X-ray Spectroscopy)
- TEM-EDX (Transmission Electron Microscope-EDX)
- Auger spectroscopy

Auger Nanoprobe at the WUSTL Image credit: Maitrayee Bose





Auger element maps of presolar silicate grain (circled) from the Acfer 094 carbonaceous chondrite

### **Structural analysis**



• TEM

FEI Themis TEM at the University of Münster



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$$E_{nuc} + E_{gr} = E_{int} + E_{rad}$$





- Stellar evolution and nucleosynthesis depend primarily on stellar mass and metallicity (other factors: mass loss, presence of companion)
- More massive stars on main sequence are more luminous, hotter, and they 'age' faster
- Nucleosynthesis occurs in stellar interiors, nucleosynthesis products are brought to stellar surface (source of matter that condenses into dust) by convection

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Low- and intermediate mass stars (M $_{\rm Init}$  < 8–10 × M $_{\odot}$ )

<u>Main sequence</u>: core H burning (p-p chain, producing <sup>4</sup>He, at higher masses the CNO cycle, producing <sup>13</sup>C, <sup>14</sup>N, and <sup>17</sup>O)

<u>RGB</u>: shell H burning (CNO cycle, NeNa cycle; main products: <u><sup>4</sup>He</u>, <sup>13</sup>C, <u><sup>14</sup>N</u>, <sup>17</sup>O, <sup>26</sup>Al)

<u>AGB</u>: shell H burning (CNO, NeNa, MgAl cycles) and He burning ( $\alpha$ captures, s-process; main products: <sup>4</sup>He, <sup>12</sup>C, <sup>16</sup>O, <sup>22</sup>Ne, n-rich isotopes, isotopes around N = 50 and 82 abundance peaks); C/O of star's envelope increases, changing the minerals condensing in the stellar winds







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**Evolutionary Tracks off the Main Sequence** perature, K

10.000



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#### Massive stars ( $M_{Init} > 8-10 \times M_{\odot}$ )

- More nuclear burning stages, than in low and intermediate mass stars, because core can heat up sufficiently enough to ignite fusion reactions of successively heavier elements (i.e., C and heavier)
- After core collapse, a large fraction of the matter overlying the core is ejected and partially and temporarily compressed/heated
- Hydrostatic and explosive burning
- Proton-, α- and n-captures, similar to what happens in low and intermediate mass stars, but at higher T and density
- Rapid n-capture = r-process, during explosion; main products are very n-rich isotopes
- *p*-process



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V1213 Cen Mróz et al. (2016)

#### Novae

 Thermonuclear runaway on a CO or a ONe white dwarf due to accumulation of H-rich matter from nearby companion (binary system)



- Explosive H burning (hot CNO cycle; main products: <sup>13</sup>C, <sup>15</sup>N, <sup>17</sup>O, <sup>22</sup>Ne, <sup>26</sup>Al)
- Heavy element isotope abundances
  unaffected

## **Presolar grain types**

SiC



Hoppe (2011) Graphite

- O-rich grains: **silicates** (olivine, enstatite, non-stoichiometric compounds), **spinel**, **corundum** (and amorphous Al<sub>2</sub>O<sub>3</sub>), hibonite, chromite, FeO, MgO, SiO<sub>2</sub>
- C-rich grains: SiC, graphite, TiC (and other refractory carbides), diamond





#### **Presolar silicates**

- In situ detection (chemical grain separation impossible)
- Abundance maximum at 200–250 ppm (by volume) in primitive meteorites and 600–700 ppm in IDPs
- Longest dimension usually 200–300 nm
- Vast majority is ferromagnesian silicates with olivine-like, enstatite-like and non-stoichiometric compositions
- Al-rich silicates, Ca-Mg-rich silicates rare





### **Presolar silicates**





- Glass (also for grains that have stoichiometric olivine or enstatite composition), single crystals of olivine and enstatite as well as polycrystalline aggregates
- Glass grains are most abundant, but sampling may be biased (e.g., grain size matters when grains are chosen for FIB lift-out and structural analysis with TEM)

Typical selected area electron diffraction (SAED) pattern of amorphous material



#### **Presolar oxides**



- Can be separated from meteorite matrices
  (but together with Solar System oxides)
- Abundance about 10–12 ppm in primitive meteorites
- Spinel, hibonite, MgO, FeO, TiO<sub>2</sub>, corundum and amorphous Al<sub>2</sub>O<sub>3</sub>
- Single crystals and crystal aggregates







- Plenty of data: <u>O</u>, Mg, Si (silicates)
- Limited/sporadic data: Ca, Ti, Cr, Fe, Ni
- Consequences of the fact that oxides can be chemically separated:
  - Oxide data are less affected by contamination than silicate data
  - Oxides have been studied for longer than silicates
- Isotope measurements of elements with low electronegativity benefited greatly from improvement of O primary ion source of NanoSIMS a few years ago (so far, silicate isotope record is most affected)



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 $10^{-1}$ 







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H burning at high T (> 40 MK) depletes
 <sup>18</sup>O, but requires extra mixing (low mass
 stars) or hot stellar envelope
 (intermediate mass stars)





 $10^{-2}$ 

(b)

<sup>17</sup>O/<sup>16</sup>O

Group 3 grains with low 17O/18O originated in low mass stars with sub-solar Z

18O/16O

<sup>18</sup>O-enrichment in some Group 3 and all Group 4 grains is best explained by nucleosynthesis in SNe

- Mg isotope data reveal more
- 3–12 % of Group 1 silicate grains have large enrichments in <sup>25</sup>Mg and small enrichments in <sup>26</sup>Mg, which is more consistent with an SN origin (nova origin unlikely for the lack of collateral shifts in O and Si isotope compositions)





 This 'SN subgroup' within Group 1 grains have so far been not recognised among oxides!





- Separation possible, but difficult
- <10 ppm in carbonaceous chondrites
- Up to several µm in diameter
- 'Onions' and 'cauliflowers'







- 'Onion'
- More abundant among high density (HD) graphite fractions
- Concentraic layers of well-graphitised carbon
- Hardly ordered assemblage of nanometre graphene sheets in core

- Cauliflower
- More abundant in low density graphite (LD) fractions
- Concentric but contorted/curved layers of graphite with no long range continuity, or
- Aggregate of 'onions'



TEM images of slices of 'onion' and 'cauliflower' presolar graphite grains from the Murchison meteorite

Croat et al. (2005)



- 'Onion'
- More abundant among HD graphite fractions
- Concentraic layers of well-graphitised carbon
- Hardly ordered assemblage of few-nm graphene sheets or TiC grain in core
- TiC inclusions enriched in Zr and Mo (refractory elements whose abundance is enhanced by the *s*-process)

- 'Cauliflower'
- More abundant in LD graphite fractions
- Concentric but contorted/curved layers of graphite with no long range continuity, or
- Aggregate of 'onions'
- TiC inclusions with Zr and Mo below detection limit, but a lot of V



TEM images of presolar graphites from the Murchison meteorite; arrows indicate TiC inclusions







- Graphite grains + Zr- and Mo-rich TiC, several times solar <sup>12</sup>C/<sup>13</sup>C, and *s*-process dominated noble gas isotope ratios probably originated in the winds of low-Z and low-mass AGB stars
- More abundant in HD graphite fractions







- Graphite grains with <sup>28</sup>Si and/or <sup>18</sup>O enriched isotope composition probably condensed in SN ejecta
- Graphite grains with evidence for the former presence of <sup>44</sup>Ti or large amounts of <sup>26</sup>Al condensed in SN ejecta
- Such grains are more common in the LD graphite fractions







• Graphite grains provide unique insight into grain condensation around evolved stars



HD graphite grains with refractory carbide inclusions







- Best studied presolar grain type
- Isotope data: Si, C, N, Ne, Mg, S, Ca, Ti, Fe, Ni, Sr, Zr, Mo, Ru, Ba
- Abundance: 30–60 ppm in the matrices of primitive meteorites
- Normally sub-µm size
- Mainstream (MS), AB (1&2), Y, Z, X, and C grains



the Murchison meteorite

#### **Presolar SiC**

Solar



-200 -400 -6001000 200 -200 0 200 400 600 800  $\delta^{30}$ Si/<sup>28</sup>Si (‰) Solar 1000 Solar 0 ۲ -1000 -1000 1000 2000 3000 0 Zinner, 2014  $\delta^{30}$ Si/<sup>28</sup>Si (‰)

200

δ<sup>29</sup>Si/<sup>28</sup>Si (‰)

§<sup>29</sup>Si/<sup>28</sup>Si (‰)

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14



- MS AB2

134 135 136 137 138

-800

-1000

84

86 87 88 92

×Sr

94 95 96 97

×Mo

98

100 130

132

×Ba

Green: s-process only, or s-process dominated isotopes





<sup>13</sup>C( $\alpha$ ,n)<sup>16</sup>O long-lasting (millenia), low n-flux (<10<sup>8</sup> ncm<sup>-3</sup>), operates at lower T (100–150MK)

 $^{22}$ Ne( $\alpha$ ,n) $^{25}$ Mg short (decades at most), high nflux (10<sup>10</sup>-10<sup>11</sup> ncm<sup>-3</sup>), operates at higher T (>300MK) only marginally activated in AGB SiC grains' parents – or is it?





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- X and C grains have peculiar isotope compositions
- SN origin likely because of C-N-Si isotope compositions, and the former presence of diagnostic shortlived radioactive isotopes (e.g., <sup>44</sup>Ti)







- X and C grains have peculiar isotope compositions
- Mixing and chemical fractionation?
- Comparison with 1D models not satisfactory







- Kinetic condensation models:
  - Molecule formation, molecular clusters, grains
  - Condensation T few 100 K
  - C/O > 1
  - Carbon dust precedes or accompanies formation of SiC in He/C zone matter





Sarangi and Cherchneff (2015)



- Presolar grain record:
- Single crystals: up to >1 μm diameter
- Polycrystalline grains: epitaxial crystal domains, twinning
- Formation of large grains not by coagulation of smaller grains (but there are exceptions)



Kodolányi et al. (2018b)

<mark>(b–f)^(g–i)</mark> = 35.5°

3C-SiC: (114)



clumpy Graphite seed

0.4

0.5

Kodolányi et al. (2018b<sub>)</sub>

- Presolar grain record:
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- Polycrystalline grains: epitaxial crystal domains, twinning
- Formation of large grains not by coagulation of smaller grains (but there are exceptions) 0.5 #1: SC13Zone5 #3: F10HeCTop #6: GB433-R02



### What comes next?



- Hyperion O ion source allows alkaline and transition metals to be measured in silicates (but isobaric interferences are an obstacle): Ca, Ti
- More, spatially better resolved data from CHILI (e.g., Mo isotopes in SiC X grains
- 3D SN nucleosynthesis models (there are some already, e.g., *Schulte et al., 2021*)
- Condensation will be better understood with a number of new TEM studies, especially the condensation of silicates





## Summary



- Presolar grains are usually small (d < 250 nm) stellar dust grains of extra Solar System origin, that are found in the matrices of primitive meteorites and in cometary dust
- Silicates, oxides, SiC, graphite are the most important
- Presolar grains carry important messages about stellar nucleosynthesis, Galactic Chemical Evolution, and grain condensation
- Most abundant presolar grain types condensed in the winds of low-mass AGB stars of about solar metallicity
- Some presolar grains come from core collapse SNe



