


October 26th to 31st
Labex **UnivEarthS**
FALL SCHOOL 2014
Villa Finaly, Florence (Italy)

COSMOS
MODELLING



Clues from the chemical and isotopic
composition of the Earth on its origin in the
Solar protoplanetary disk

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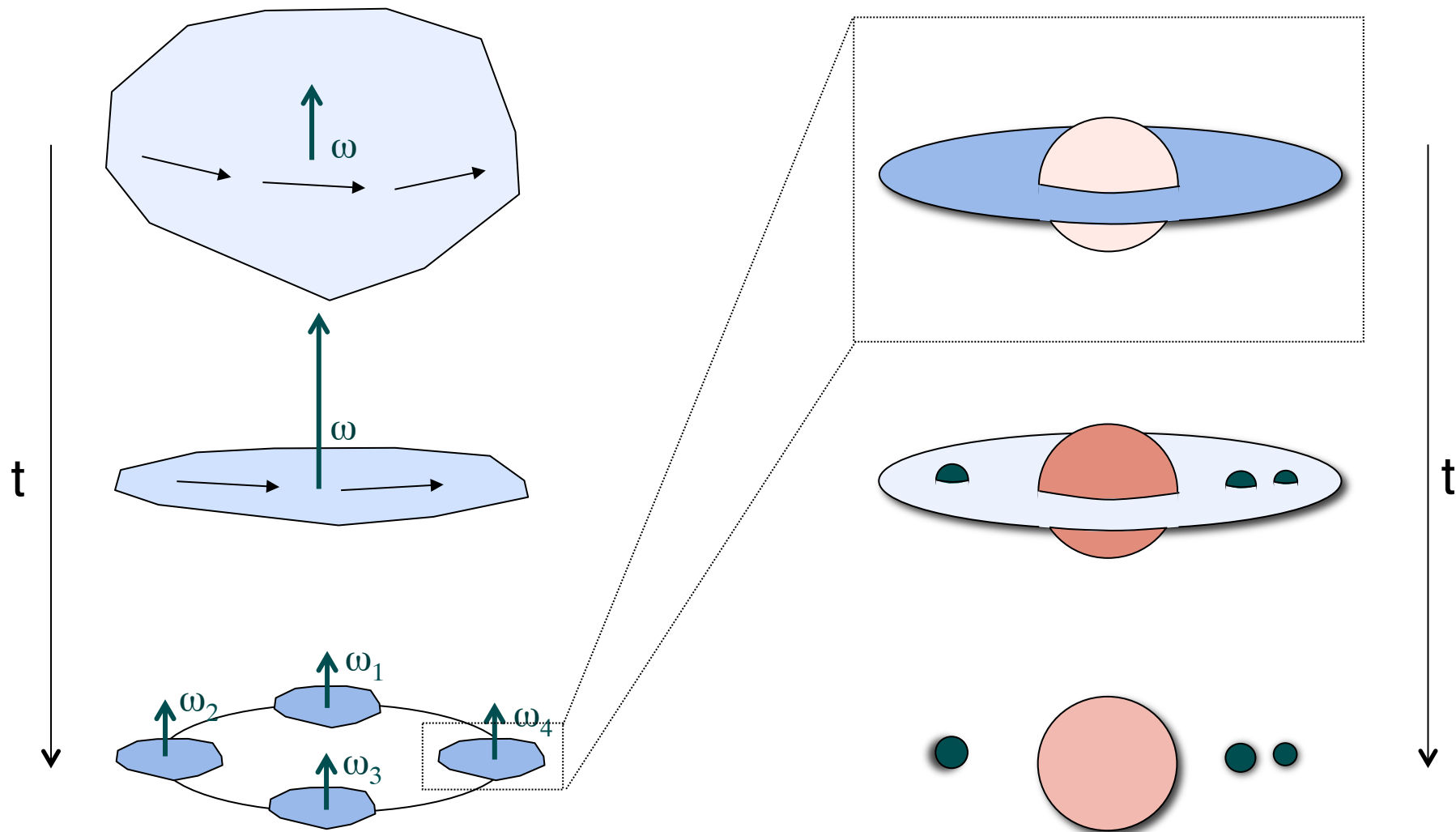
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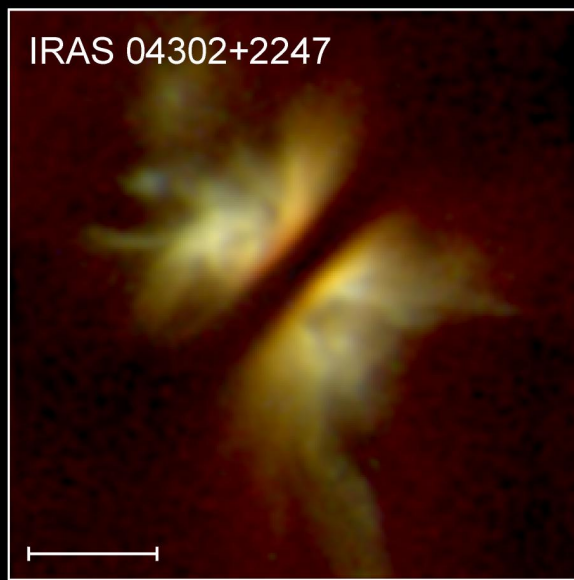
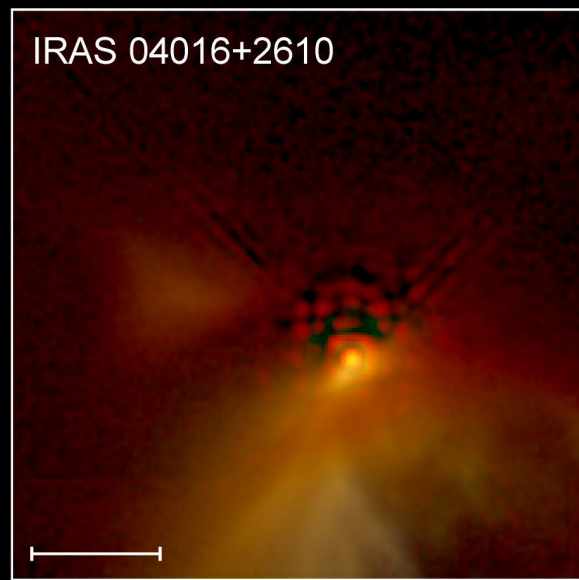
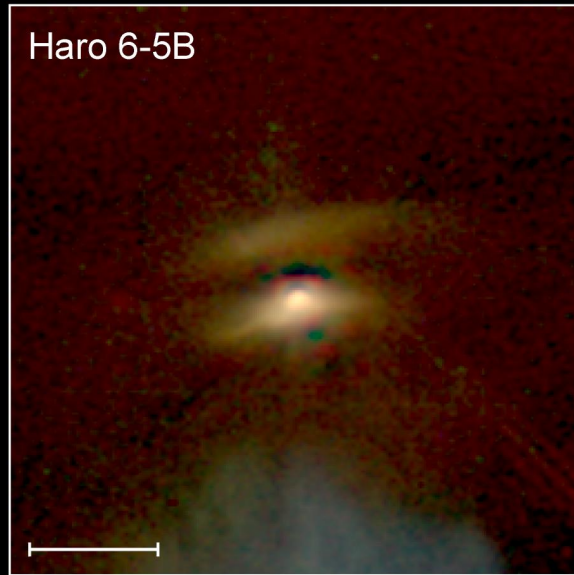
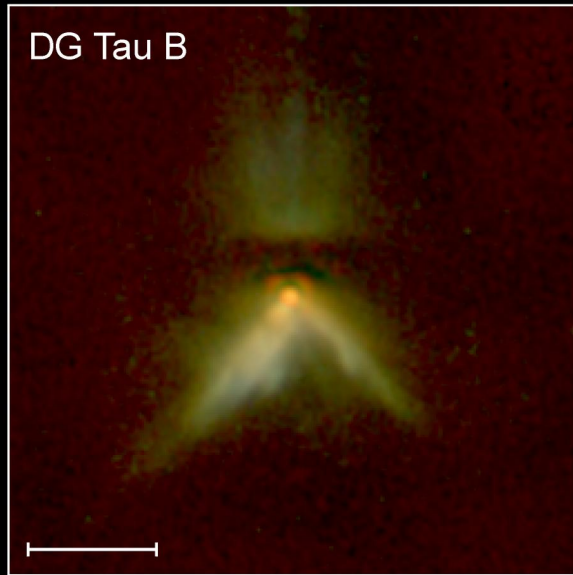
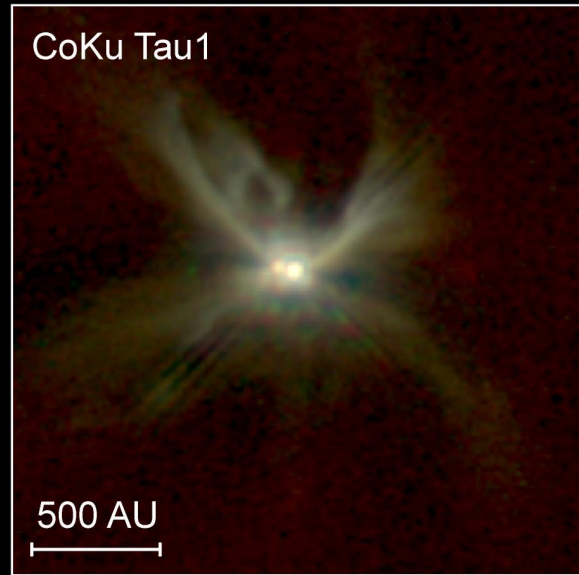
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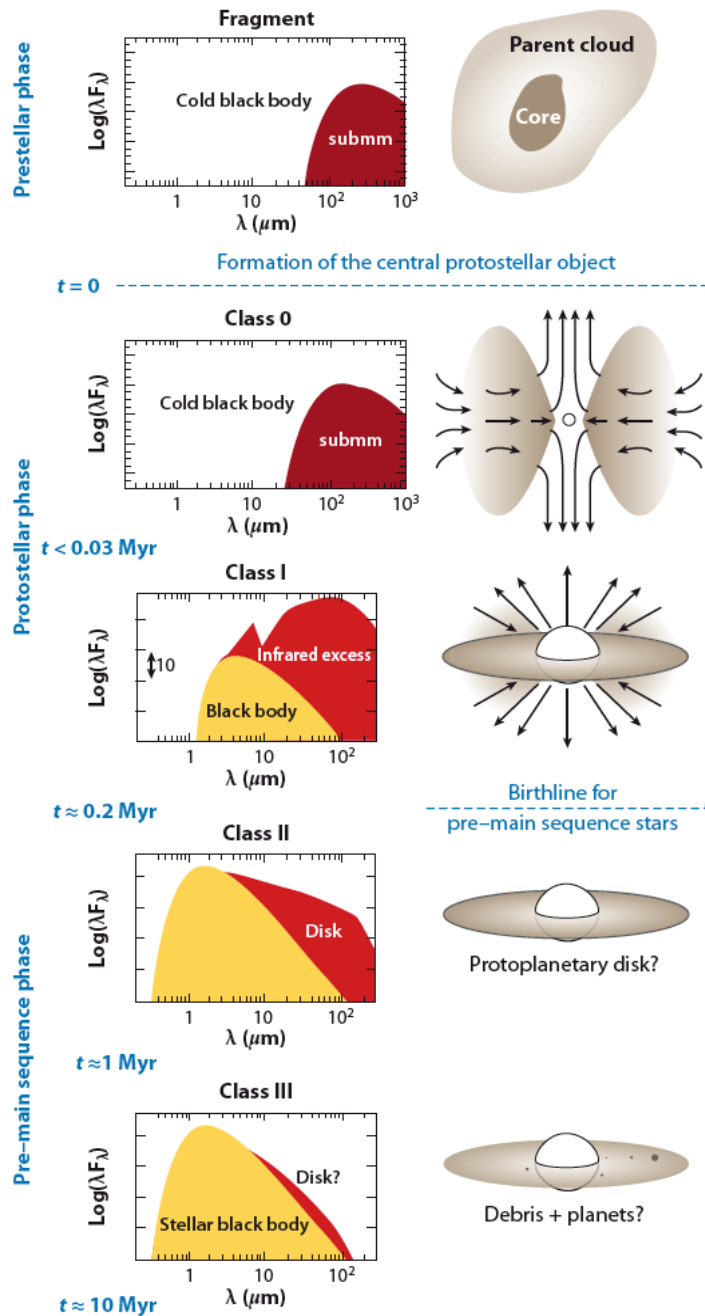




Adapted from Kutner 2007



HST images, Taurus-Auriga molecular cloud (Padget et al., 1999)



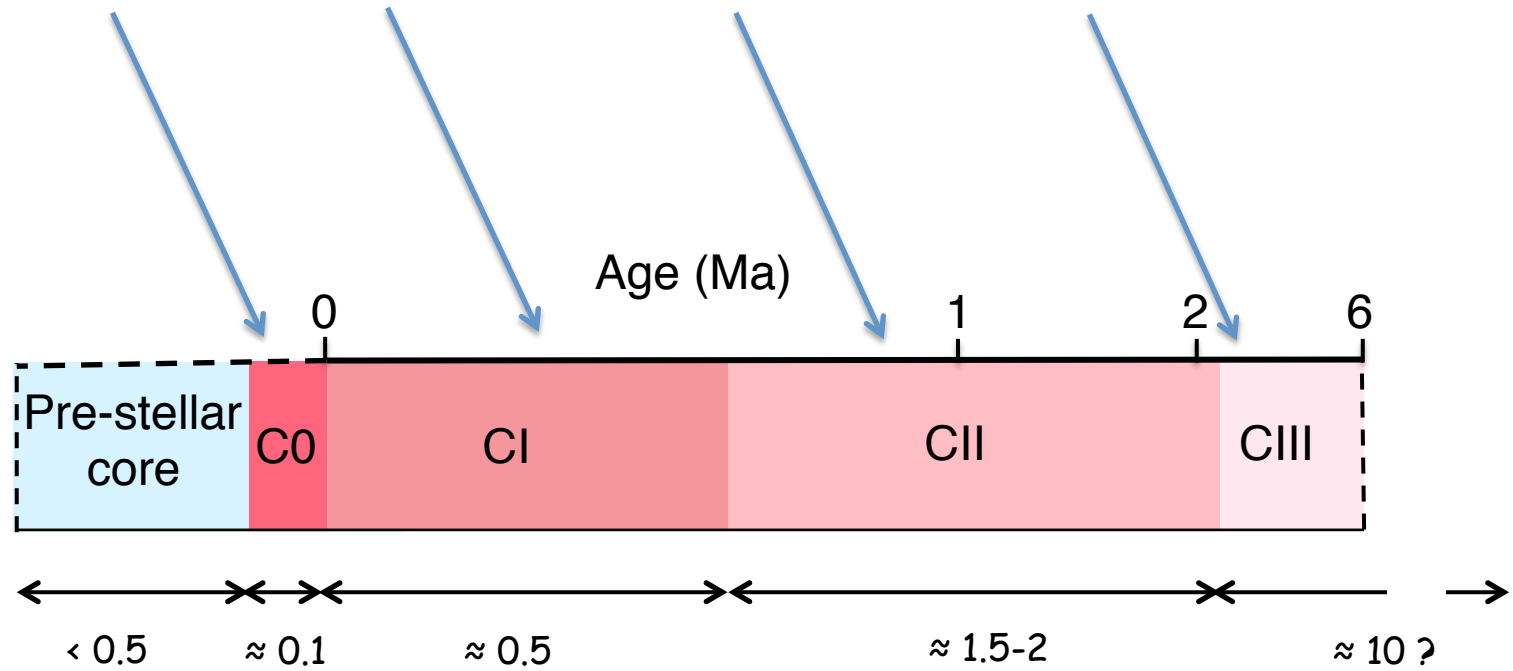
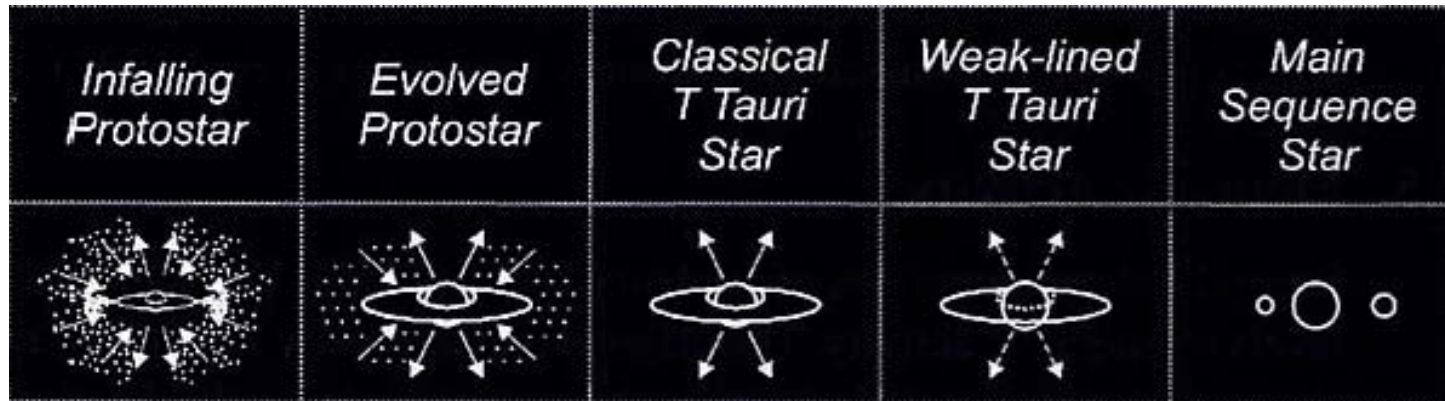
Variations in the spectral energy distribution (SED) from class 0, I, II to III

Infall

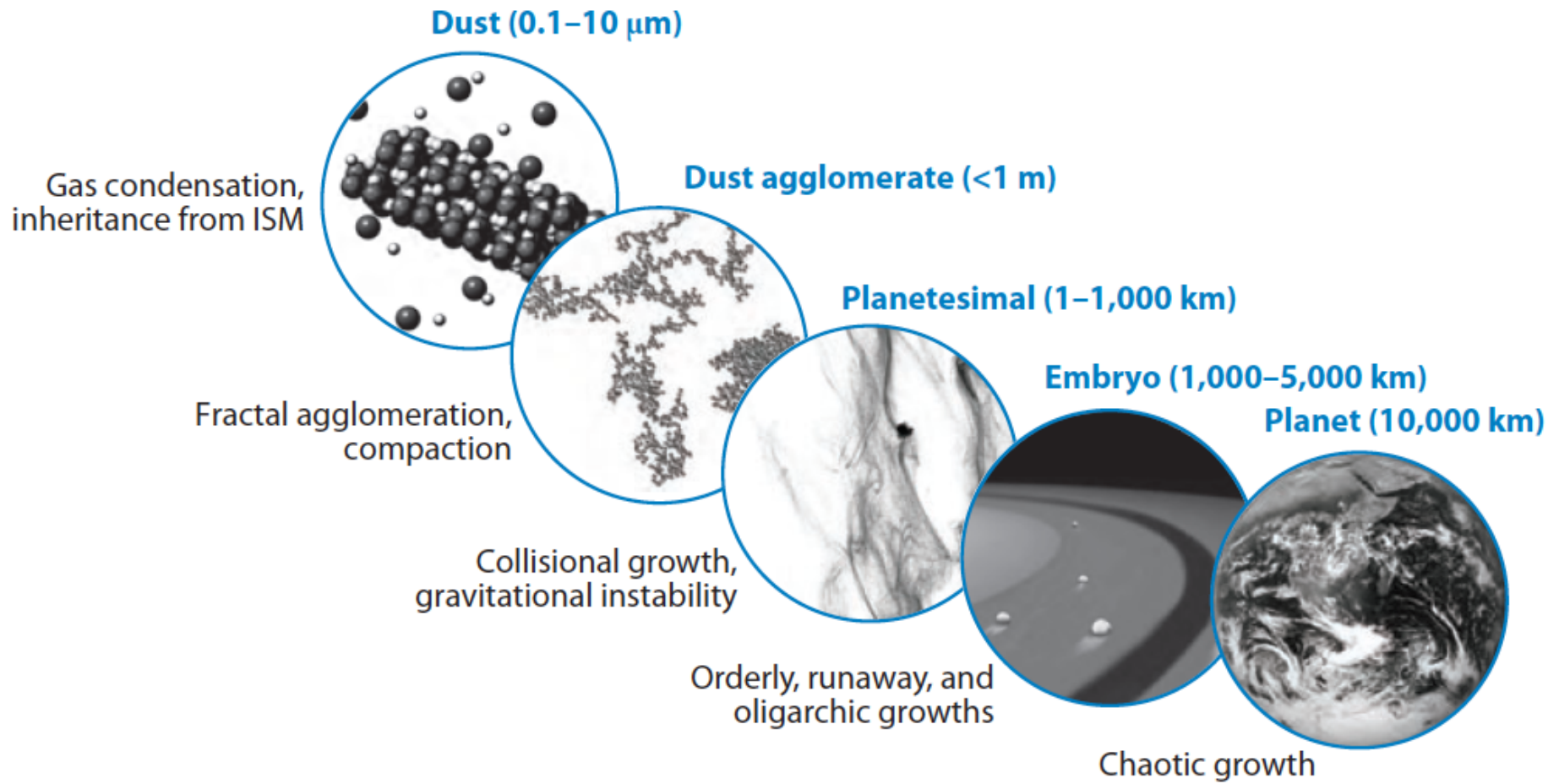
Accretion of the star from its disk

Dissipation of the disk

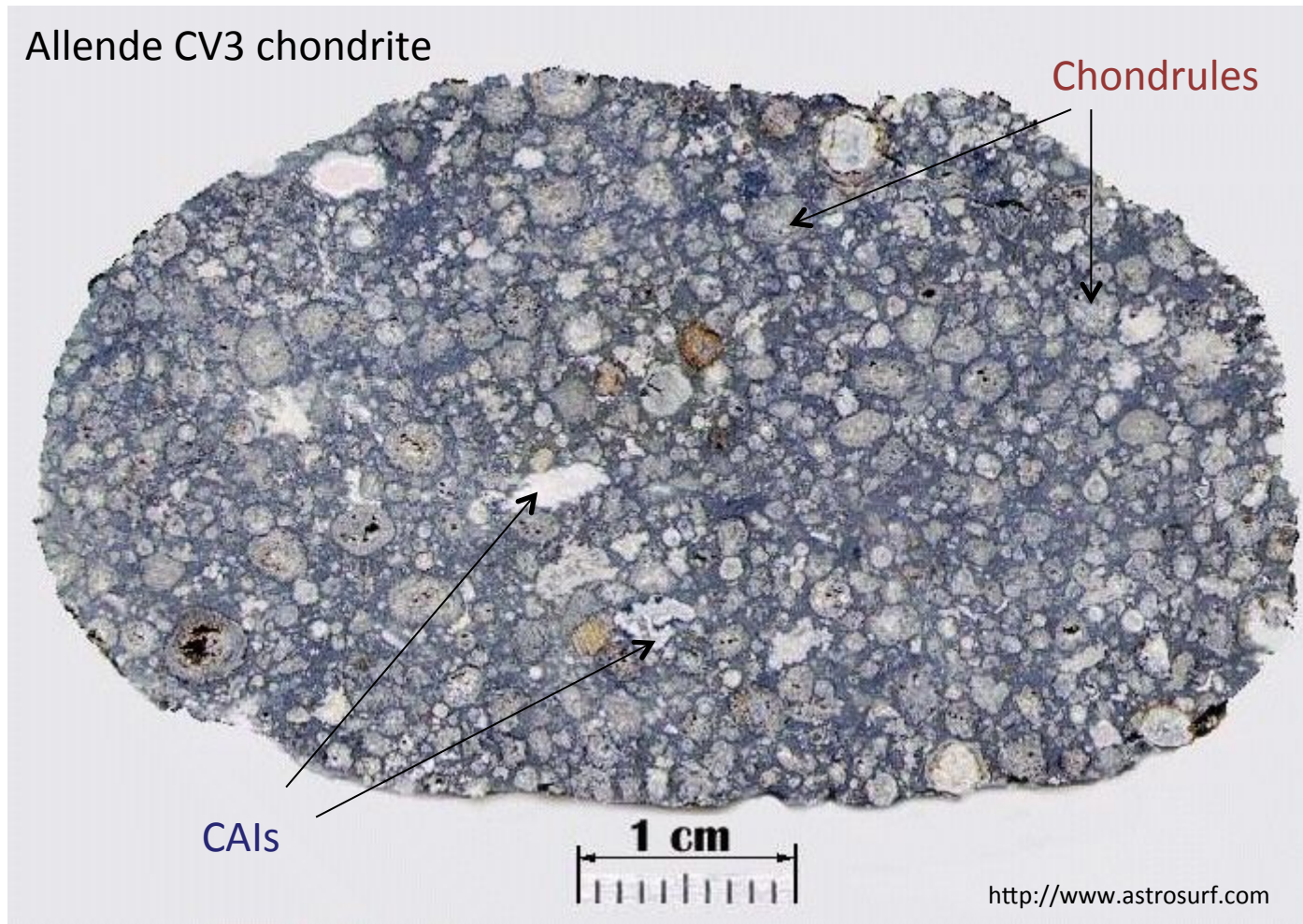
Classification from André & Montmerle (1994)



Time scale (Ma) from Evans et al. (2009)



Chondritic meteorites are understood as fragments of small planets accreted from the earliest solids formed in the disk: they are “sediments” made of two major high-temperature components: Ca-, Al-rich inclusions (CAIs) and chondrules



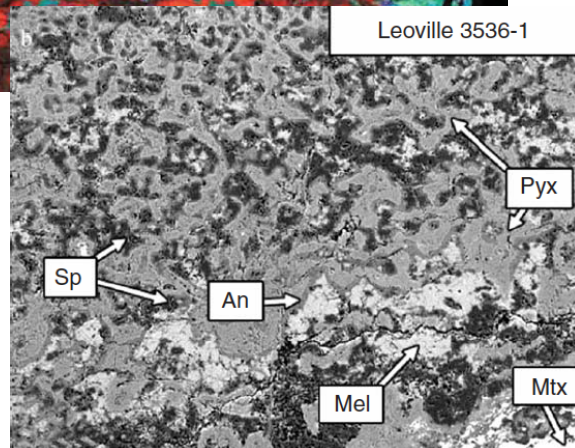
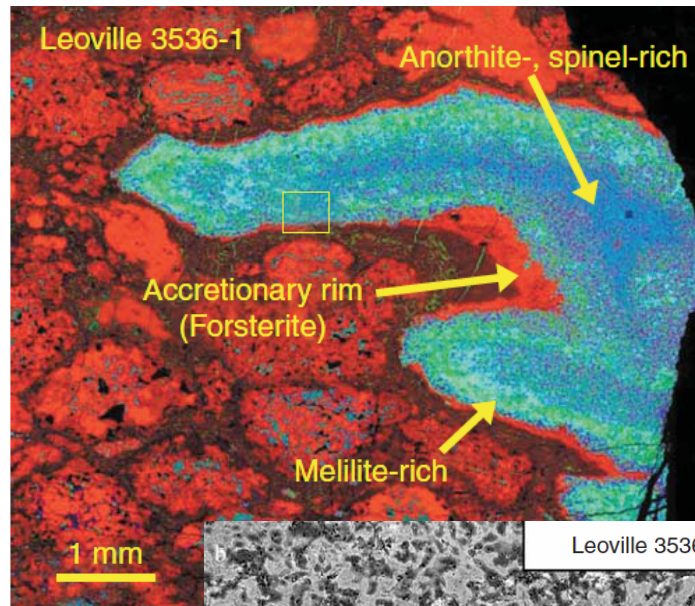
The two high-temperature components of chondrites

Refractory objects:

(e.g. Ca-, Al-rich inclusions, CAIs)

Show traces of condensation/evaporation, accretion in the gas, melting and re-melting in the gas

ORIGIN: formed from precursors issued from condensation



Fine grained spinel-rich CAI
Mac Pherson 2005

Chondrules

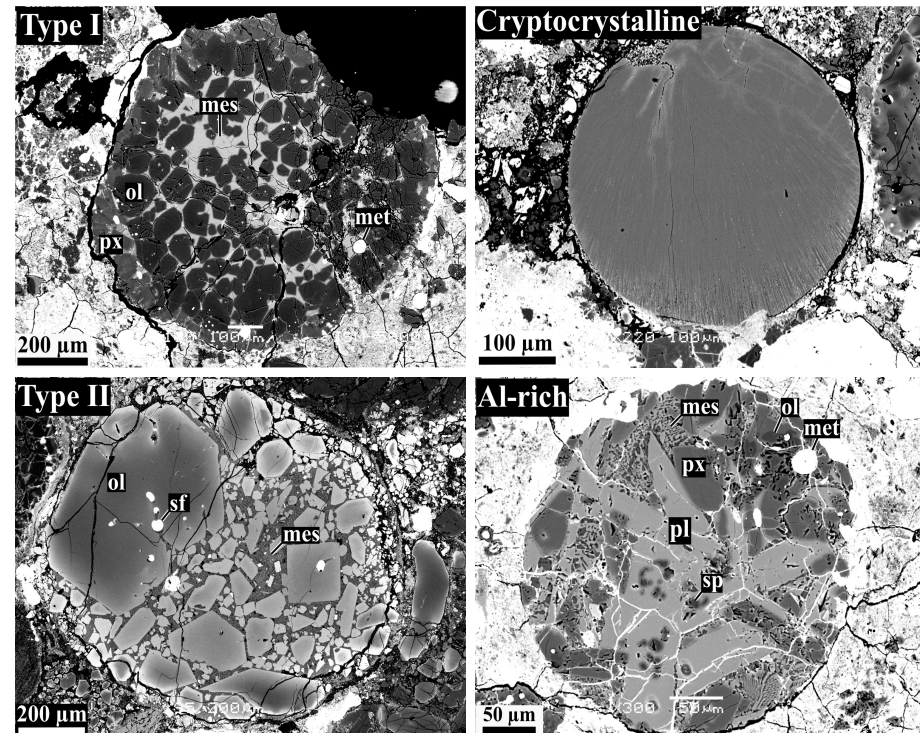
(e. g. ferromagnesian porphyritic chondrules)

Less refractory, silicate spherules once molten

(totally or partially in the gas),

40-80 % by volume of most chondrites

ORIGIN: unknown (energetic events, shock waves, collisions between planetesimals, ...)



Courtesy Sasha Krot

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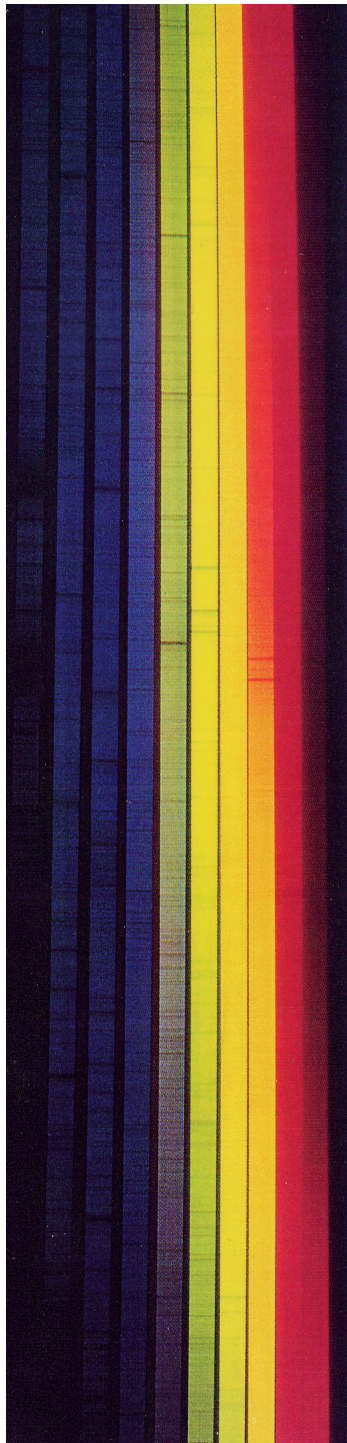
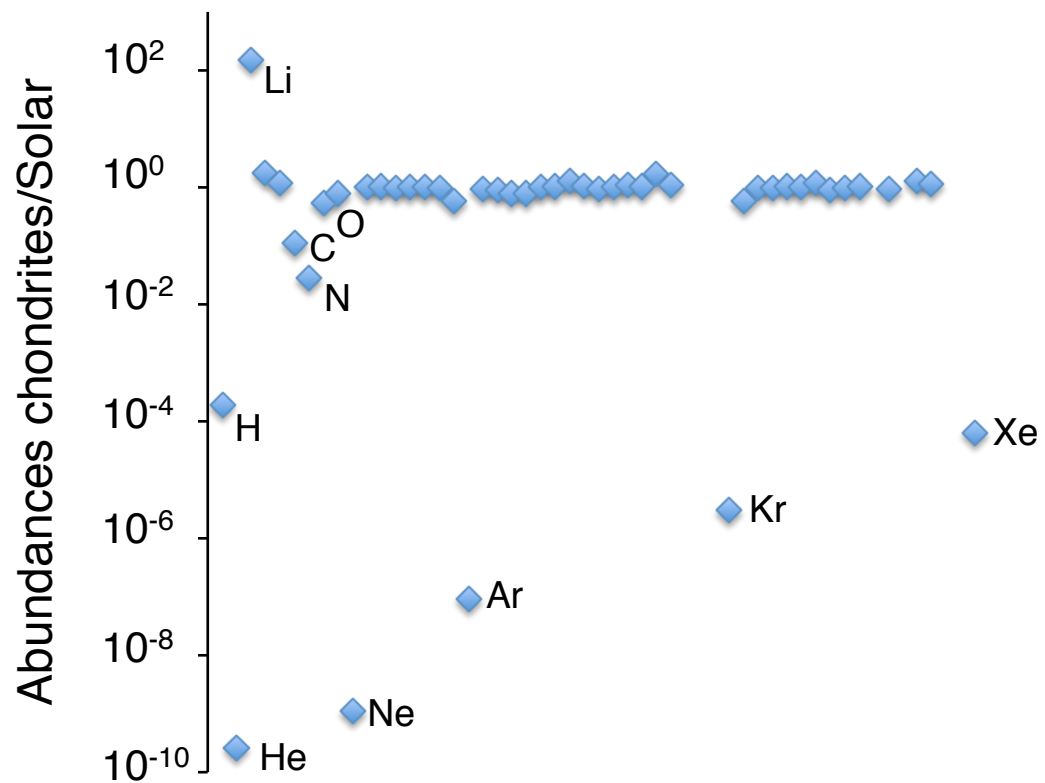
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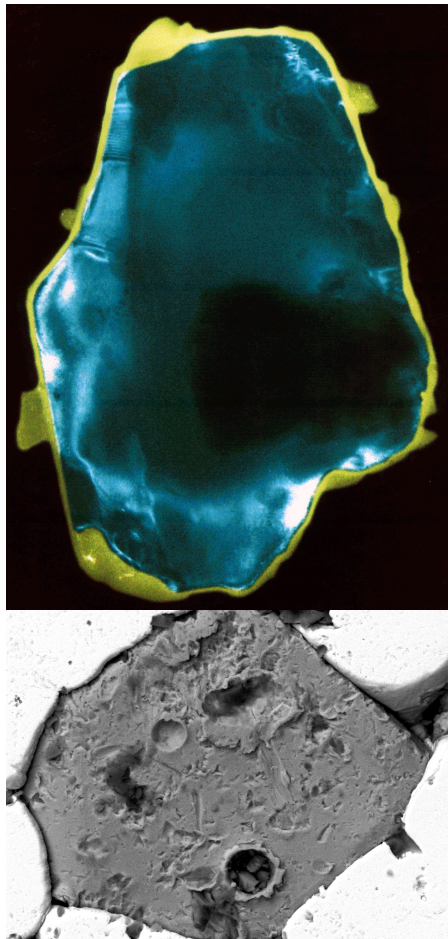
Primitive (non-differentiated) meteorites have the same composition than the Solar photosphere (i.e. the gas of the SPD) except for gases (H+rare gases) and also Li, N, C & O



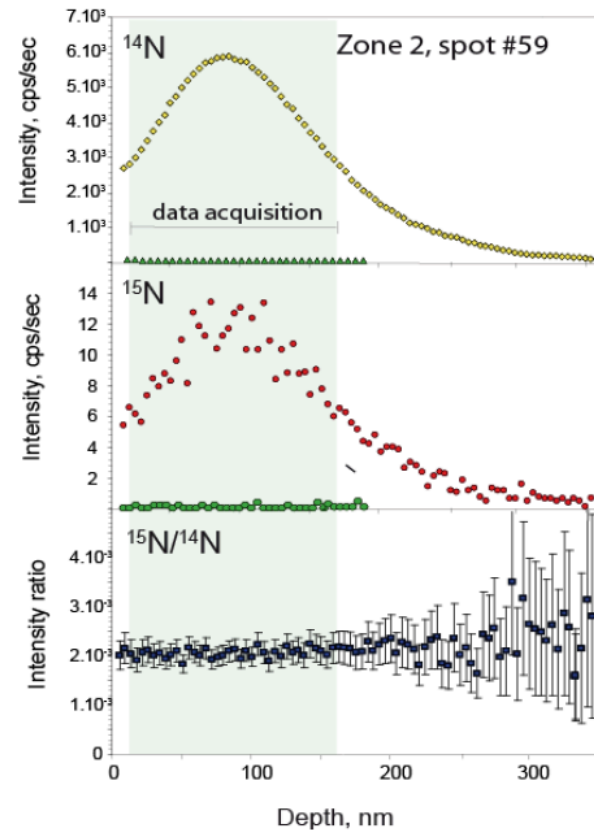
The isotopic composition of the Sun is (presumably) similar to chondrites except for:

- H (D burning during accretion of the Sun)
- Li (${}^7\text{Li}$ selective burning relative to ${}^6\text{Li}$ in the convective zone)
- O (enrichment of the Sun in ${}^{16}\text{O}$: origin still controversial and not well understood)
- N (enrichment of the Sun in ${}^{14}\text{N}$: origin still controversial and not well understood)
- C (enrichment of the Sun in ${}^{12}\text{C}$: origin still controversial and not well understood)

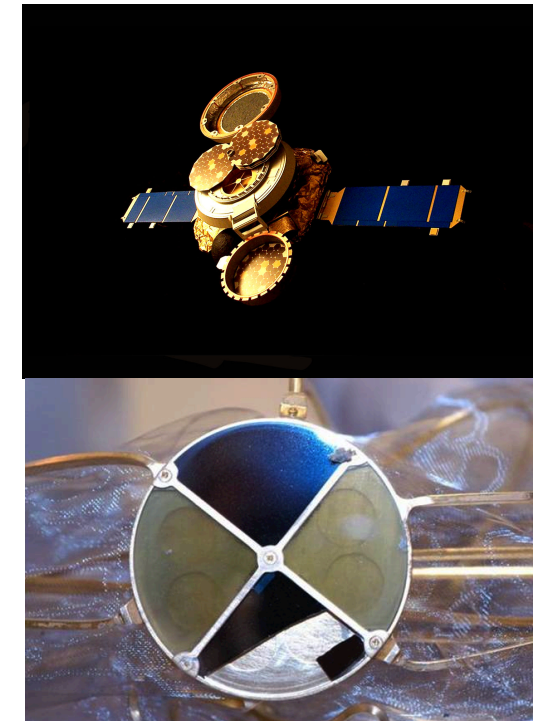
Lunar soils



Ion microprobe isotopic depth profiles

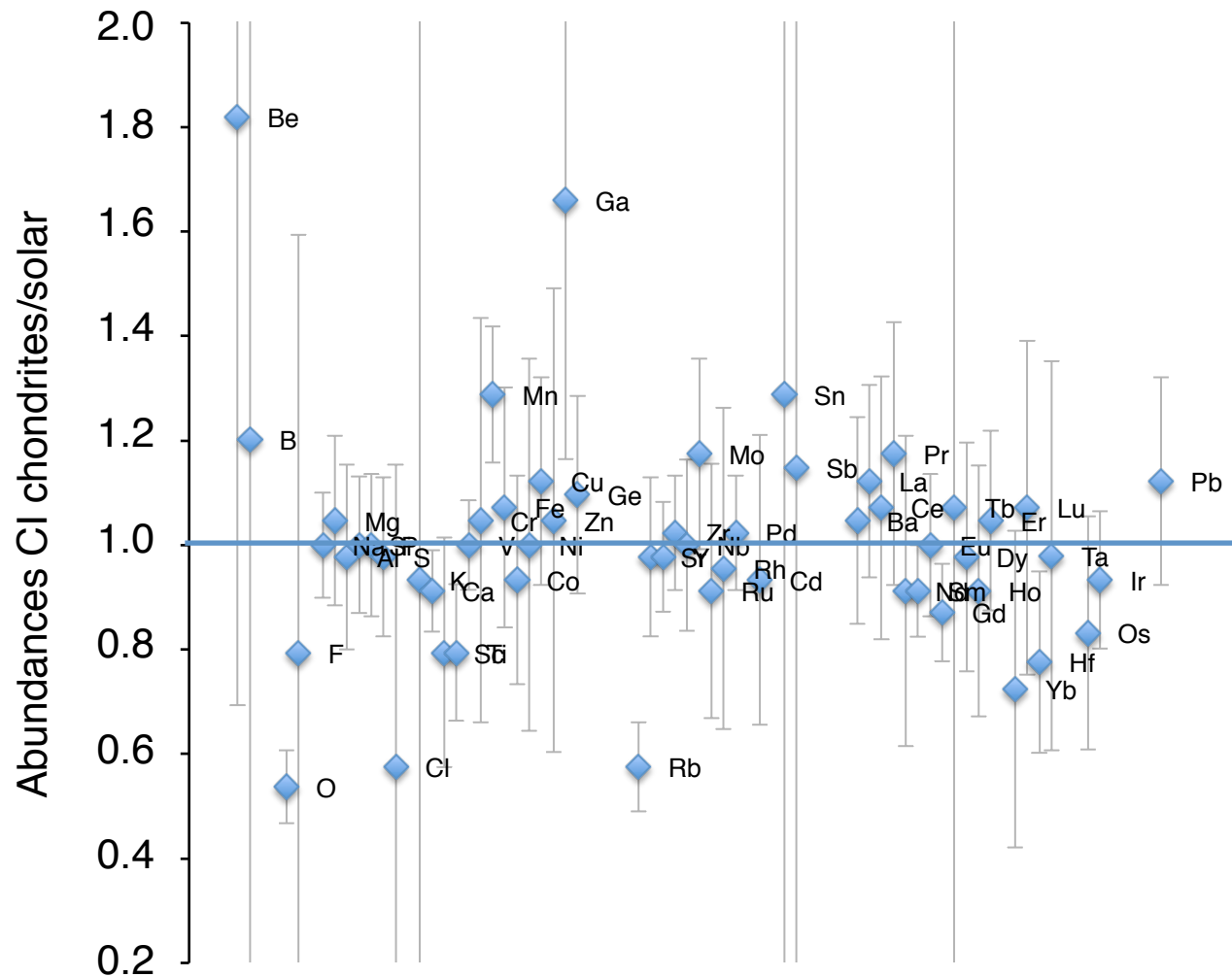


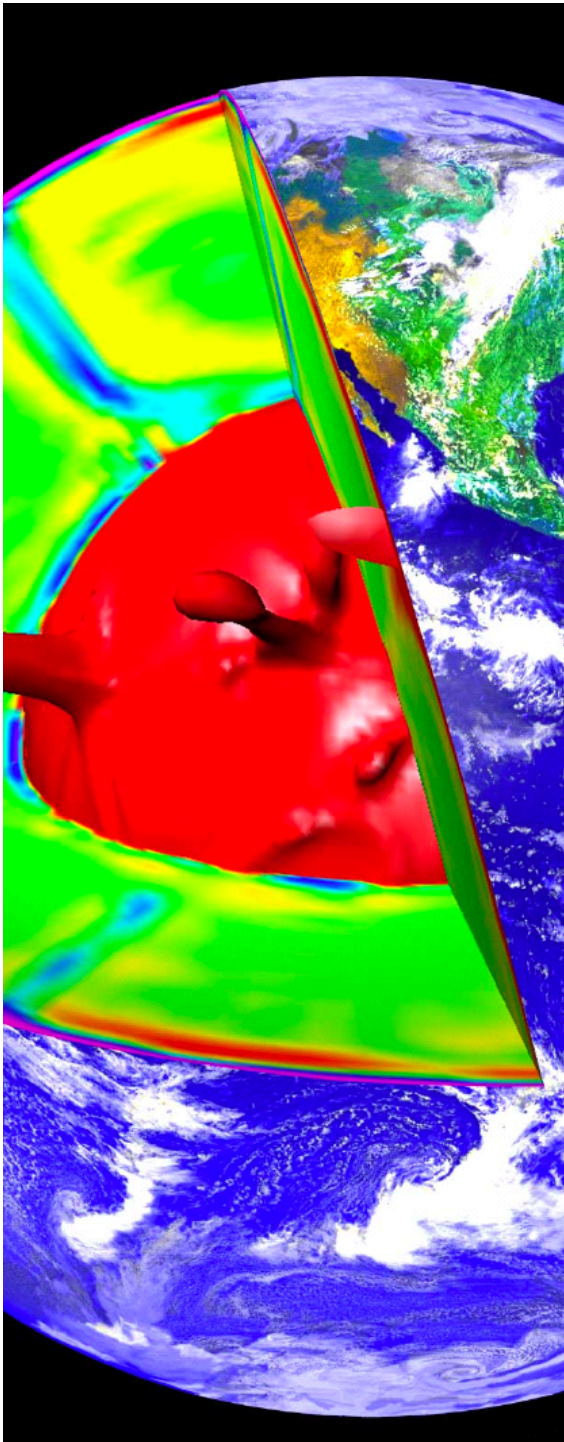
Genesis collectors



Chaussidon & Robert *Nature* 1999; Hashizume, Chaussidon Marty & Robert *Science* 2000; Hashizume, Chaussidon, Marty & Terada *ApJ* 2004; Hashizume & Chaussidon *Nature* 2005; Hashizume & Chaussidon *GCA* 2009; Marty, Chaussidon, Wiens, Jurewicz & Burnett *Science* 2011.

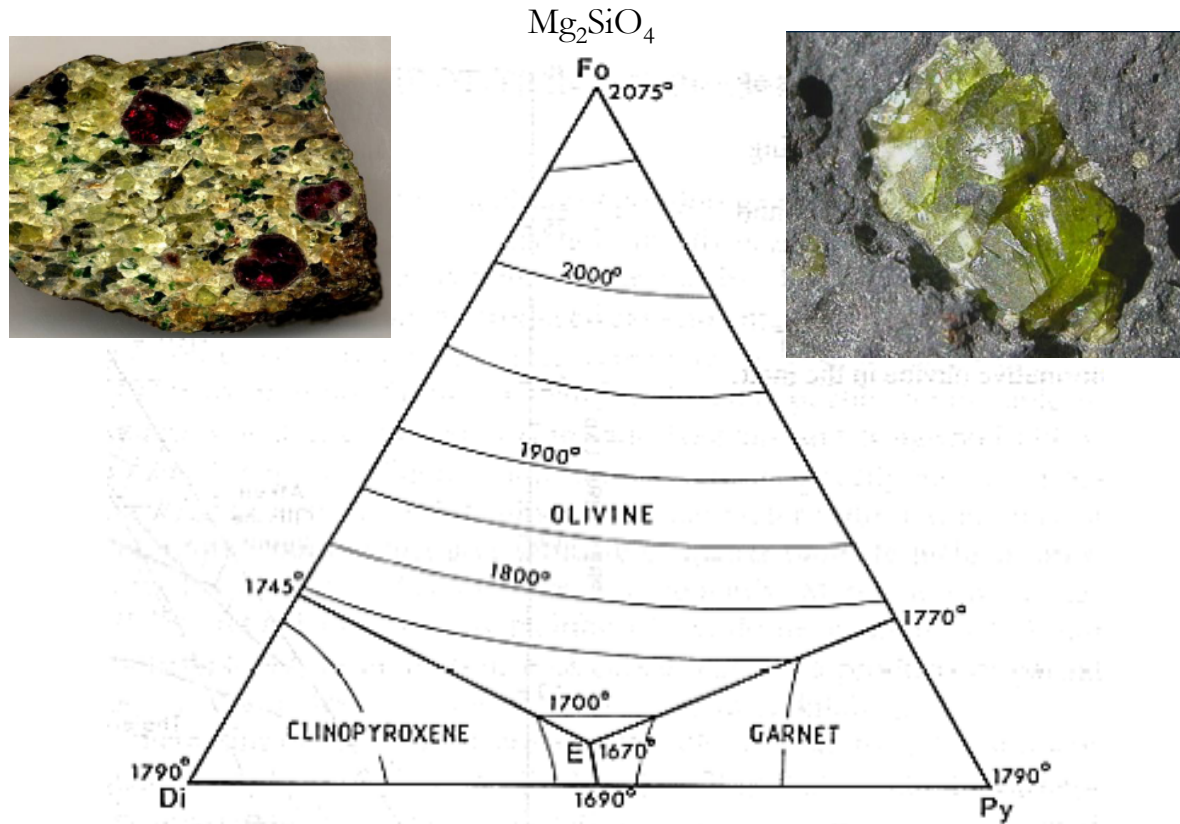
The agreement is nearly “perfect” for all elements
(otherwise probably error in measurement of concentration in the Solar photosphere)



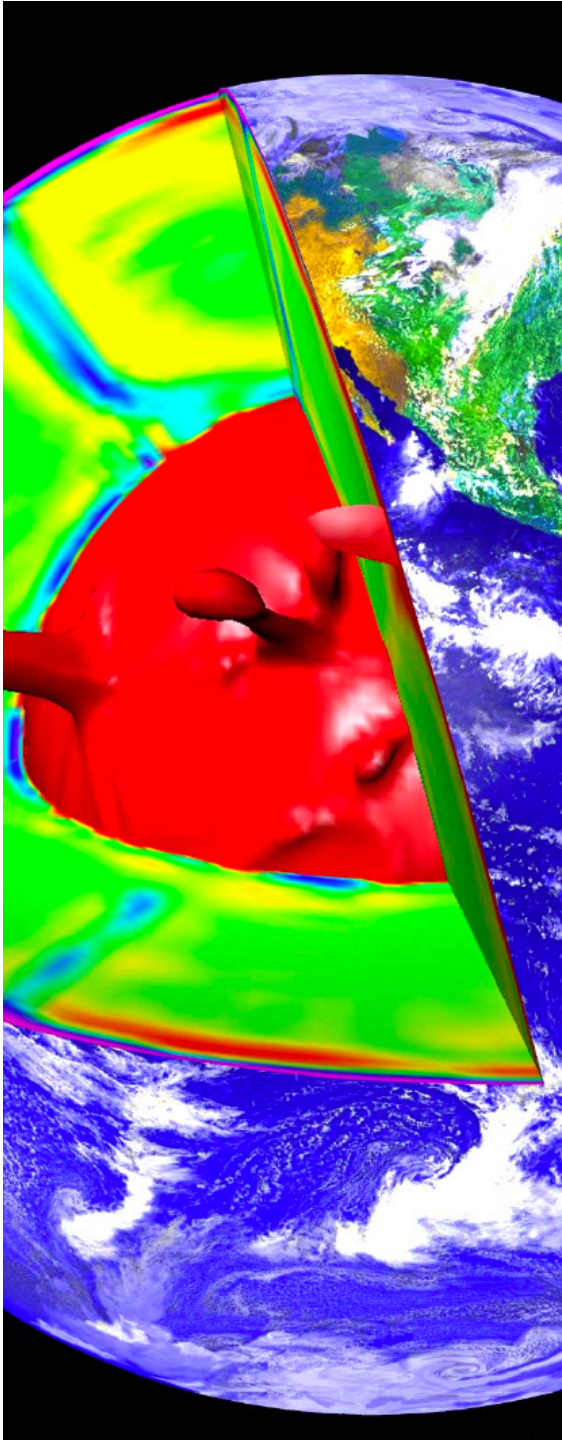


How to determine to first order the composition of the bulk Earth (crust+mantle+core) for major elements independently of meteorites and with robust assumptions (e.g. Allègre et al., 1995) ?

- use the composition of upper mantle rocks (fertile peridotites)

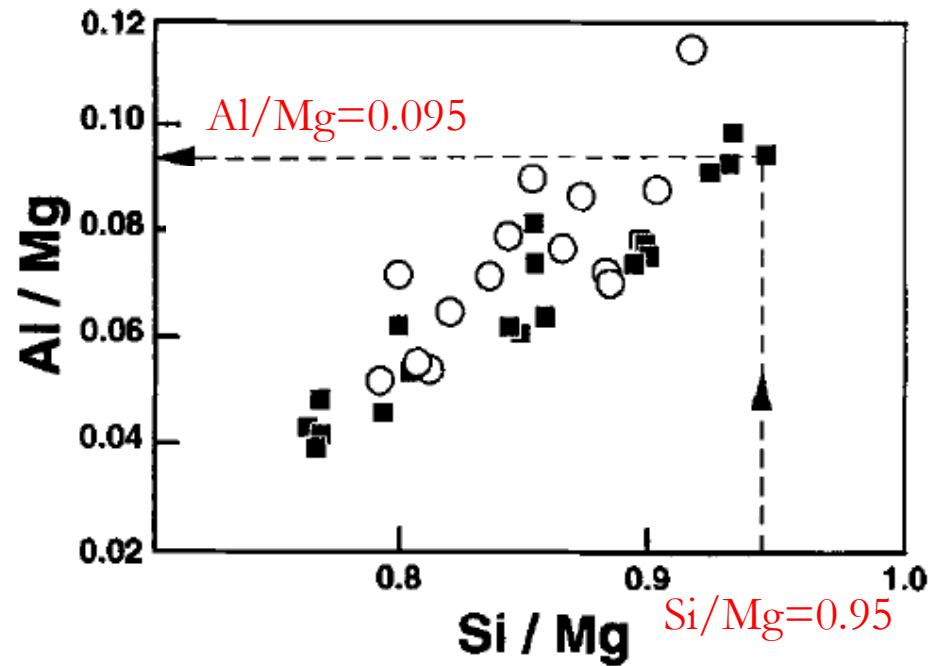


Example of experimental phase relationships for melting of peridotite (without Fe) at 40 kb (curves are liquidus)



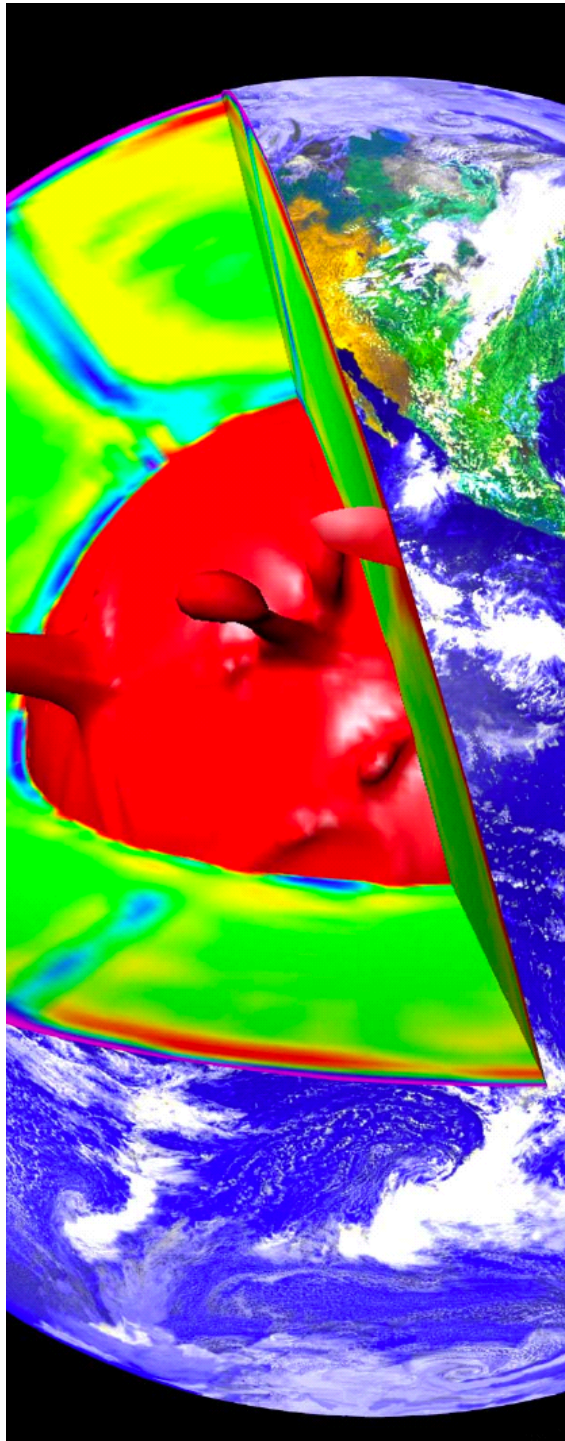
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- use the composition of upper mantle rocks (fertile peridotites)
- assume that the bulk mantle \approx upper mantle (proportion of crust = 0.5% total silicate Earth)
- use ratios of lithophile elements (Al, Ca, Mg, Si)



All ratios are at \pm a few percent relative

Ca/Al=1.24 (peridotites), =1.07 (chondrites)
Mg/(Mg+Fe)= 0.9 (peridotites), thus Fe/Al=2.68



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- assume a core having $\approx 32\%$ mass of the Earth made of Fe and Ni ($\approx 6\text{ wt}\%$) plus $\approx 8\text{-}10\%$ of light elements?
- problems with Si (how much Si in the core? S and O with Si)

Si 17.2 wt%

Al 1.5 wt%

Mg 15.9 wt%

Ca 1.6 wt%

Fe 28.2 wt%

O 32.4 wt%

= CMAS (Ca+Mg+Al+Si) + Fe + O

+ minor (Na+K+Mn+Ti+Ni + Cr $\approx 3\text{ wt}\%$) and trace elements

The bulk Earth is depleted in volatile elements (elements having low condensation temperatures).

The ratio to Si of refractory elements is \approx chondritic

(depletion in O relative to CI explains the 1.4 enrichment in all refractory elements)

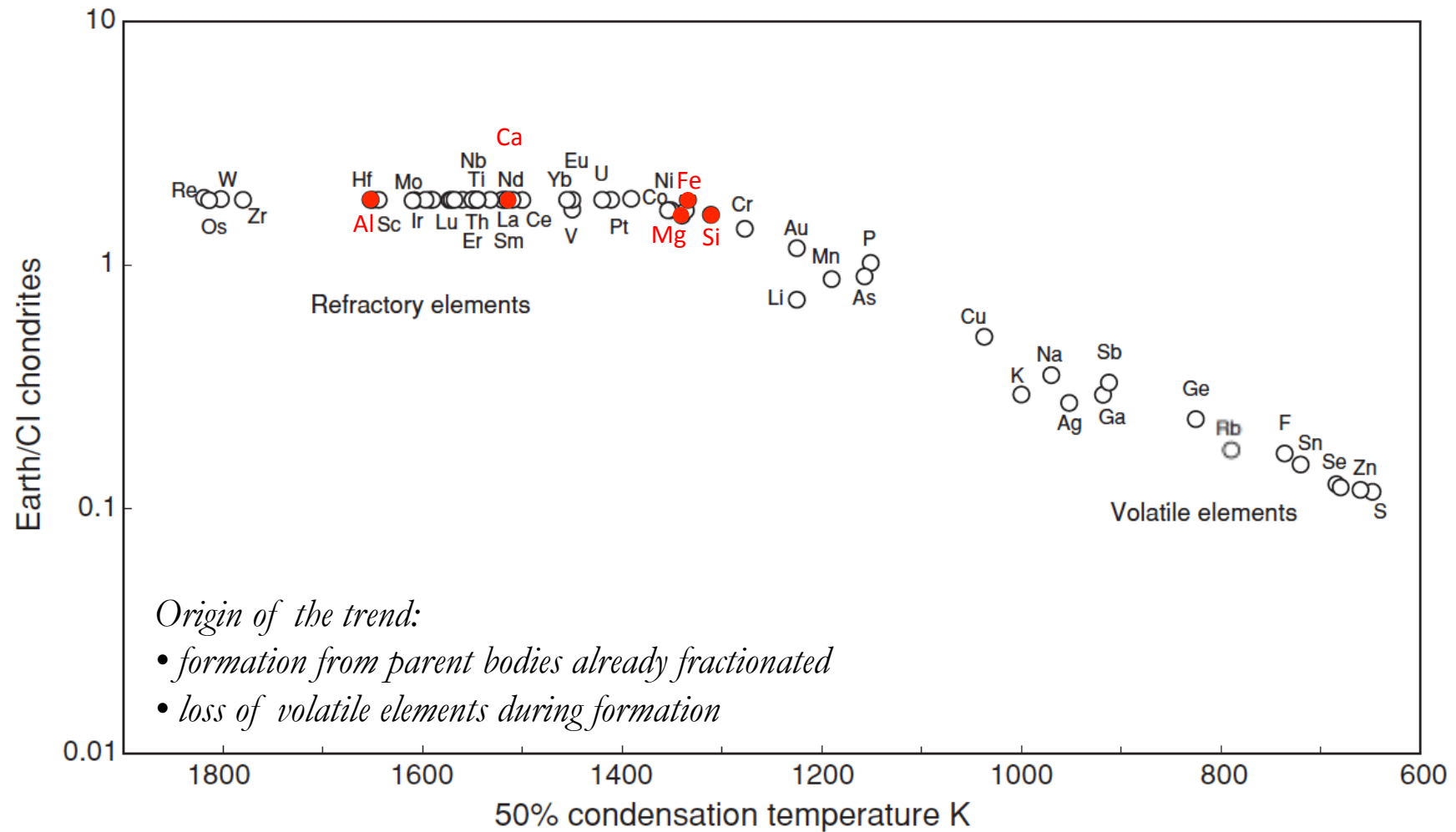
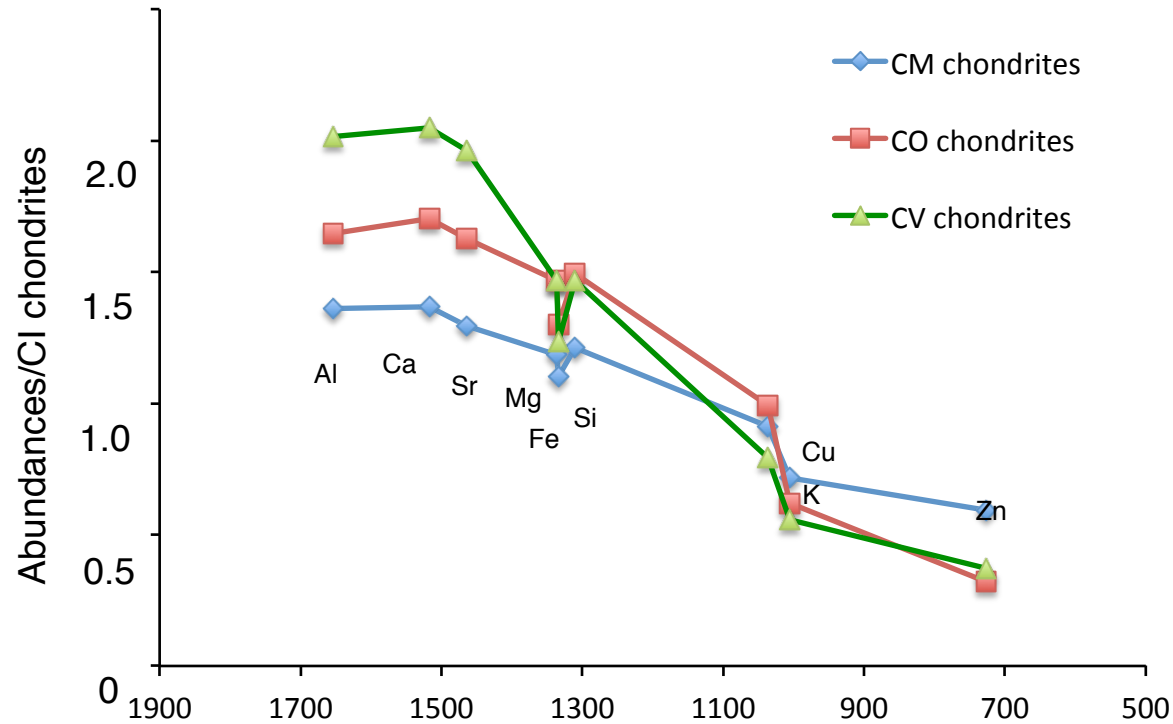


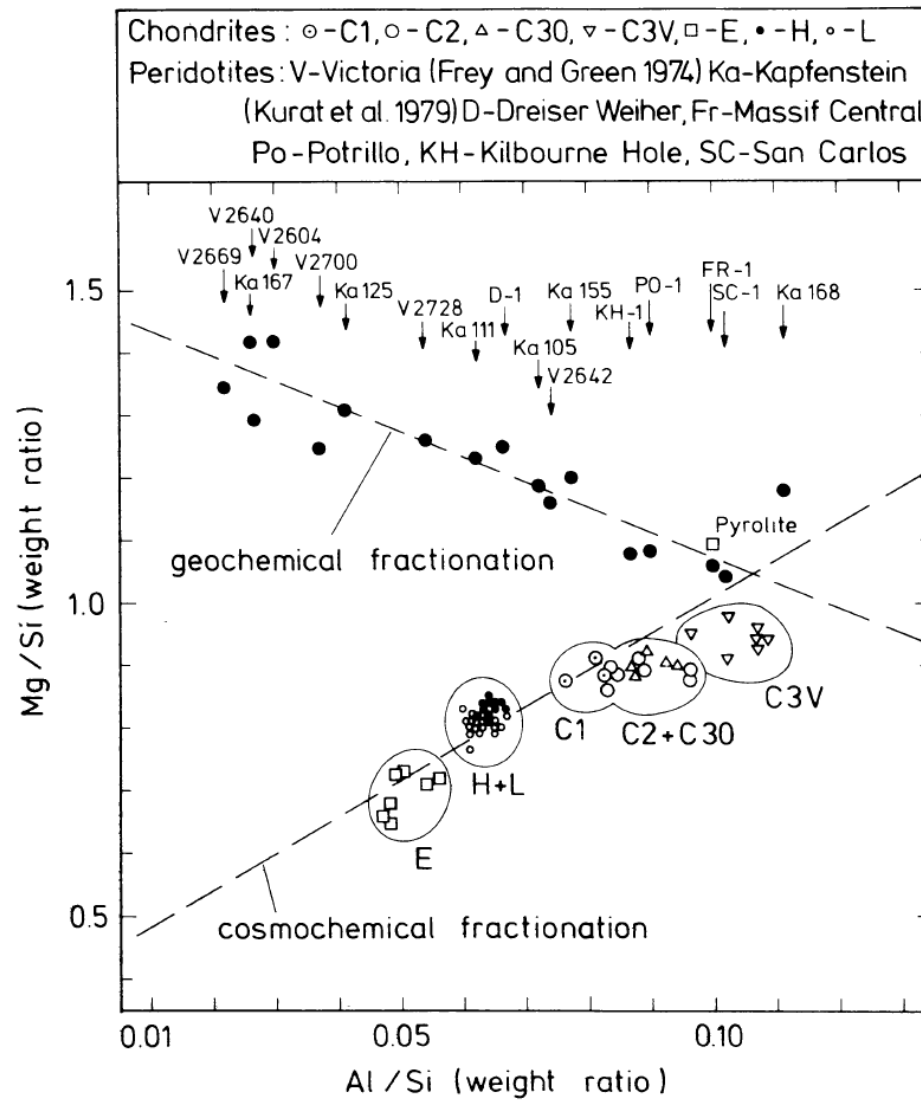
Fig from Albarède 2008

Effects of fractionation related to condensation temperature are also observed among chondrites (though primitive, i.e non-differentiated)



The systematics shown by chondrites is called the cosmochemical trend: it must reflect processes which fractionated refractory/ volatile elements during the formation of the constituents of chondrites (CAIs, chondrules, matrix) before their accretion

To first order, the bulk composition of the Earth is understood as reflecting its formation from chondritic parent bodies with variations in composition due to fractionation processes driven by the volatility of the elements



Note that the intersection implies that several wt% Si are in the core

Jagoutz et al., 1979

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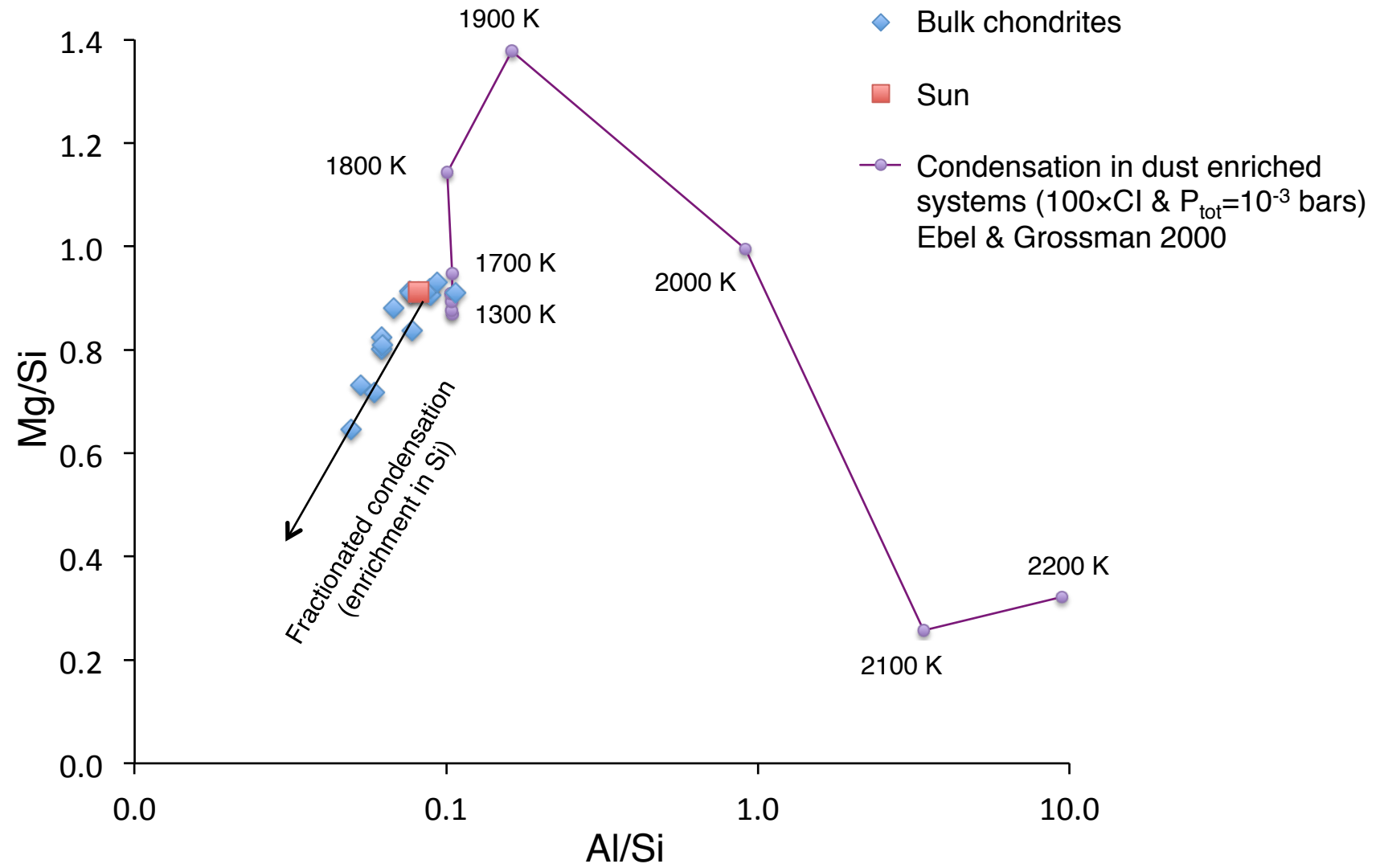
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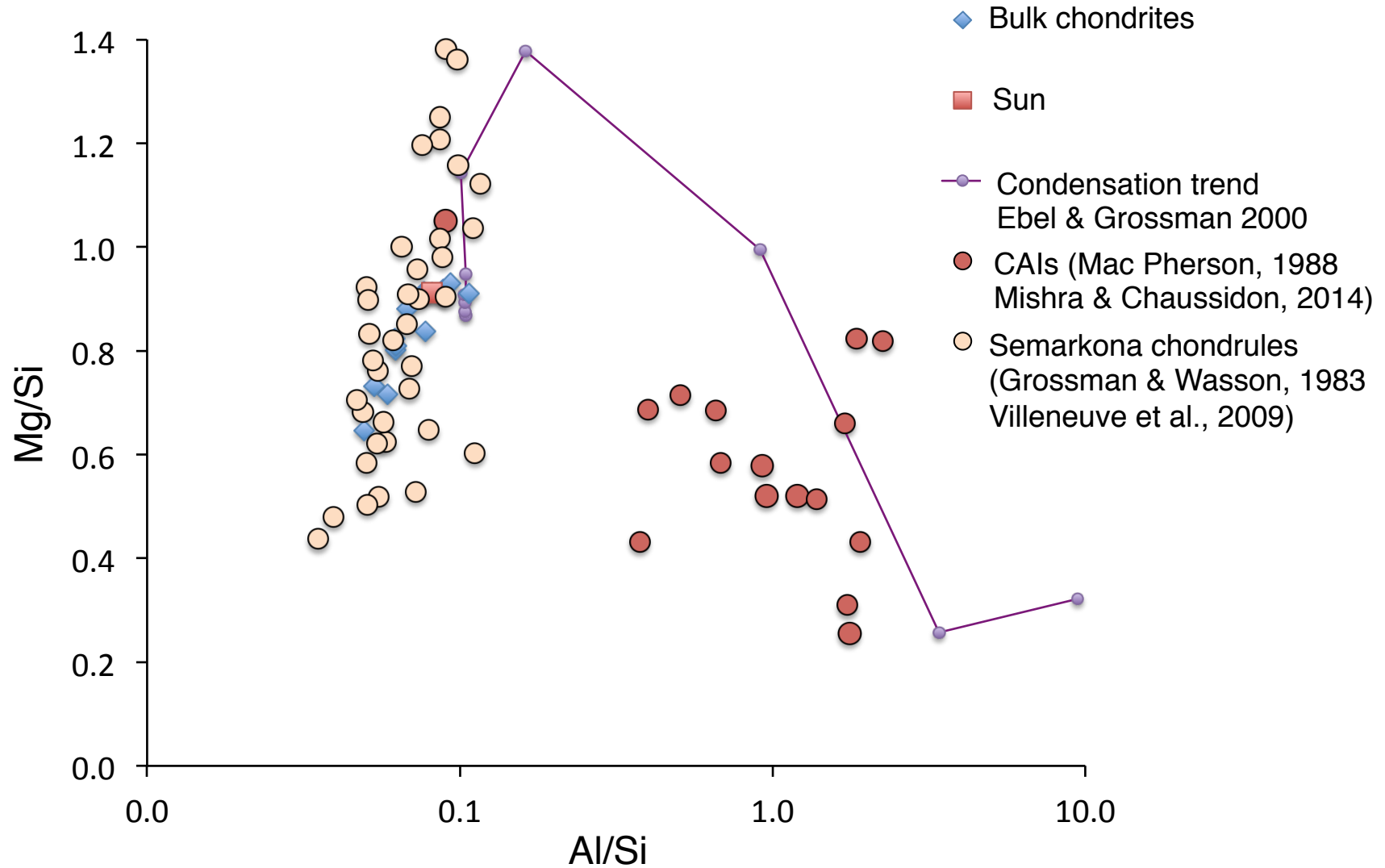
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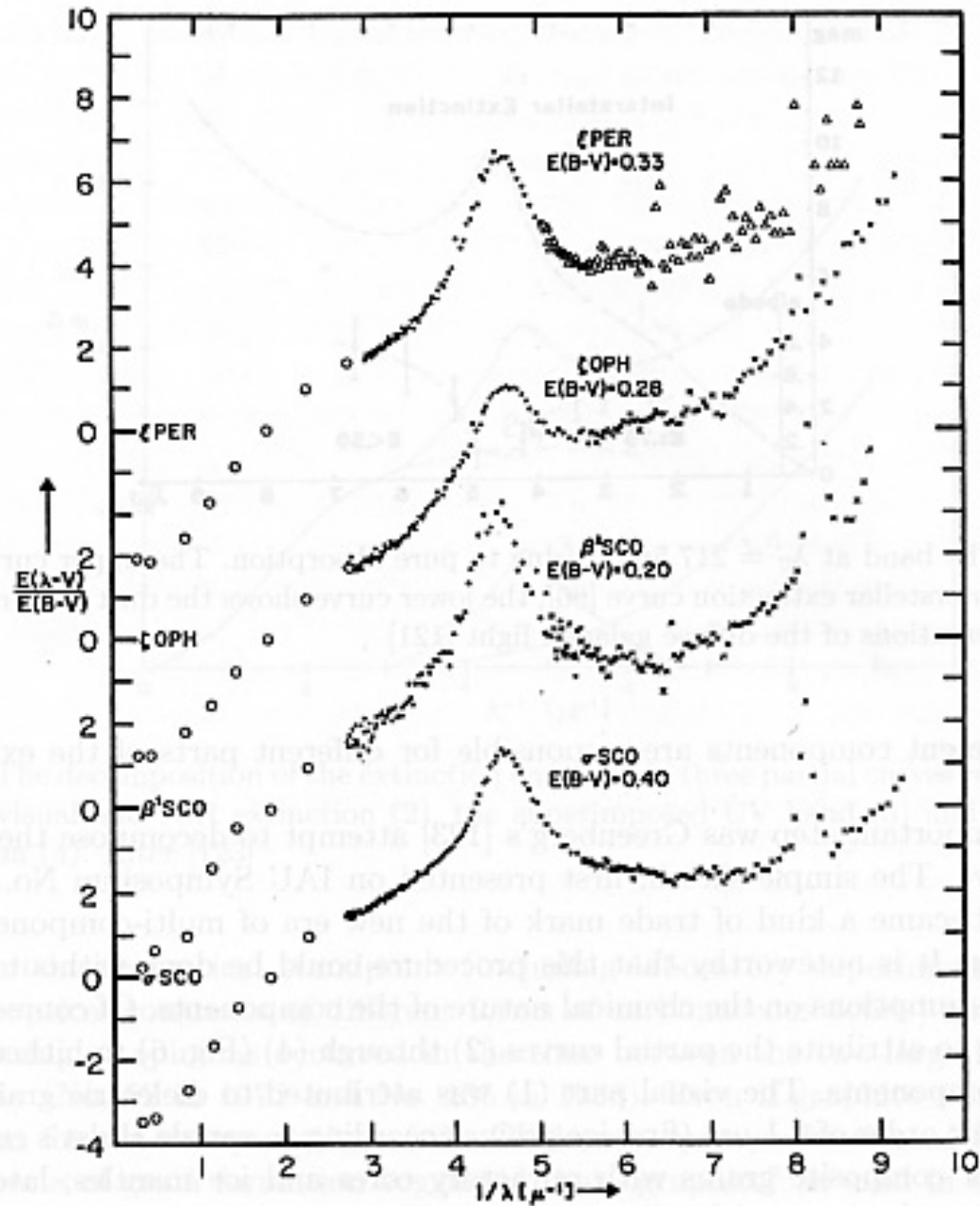
The bulk composition of chondrites reflect various proportions of CAIs and chondrules (and matrix) condensed under various conditions



What is the origin of the solids in the Solar System?

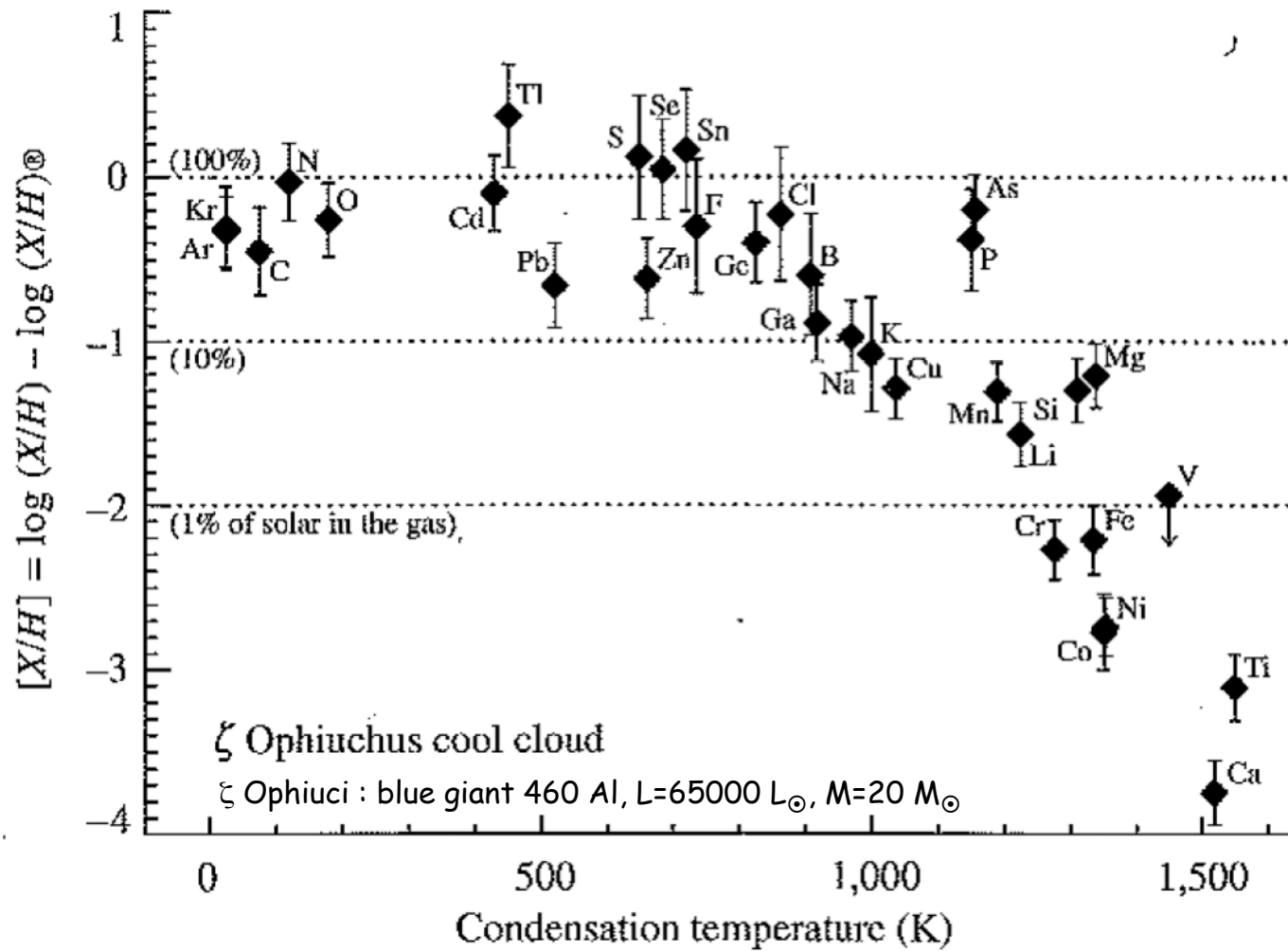
Why do all models consider that all (nearly all) of the dust in the Solar protoplanetary disk was formed by condensation in the disk from the Solar nebula gas?

- No definitive clues from astrophysical observations:
 - what would the interstellar dust look like?
 - observations of disks
- No definitive clues from astrophysical models
 - coupling models of the infall of the parent molecular cloud with models of the disk have never been done
- Constraints from observations in meteorites
 - presolar grains with isotopic anomalies
 - the question of the organics
 - observation in primitive meteorites of condensation products having Solar isotopic composition and traces of isotopic fractionation in the SPD

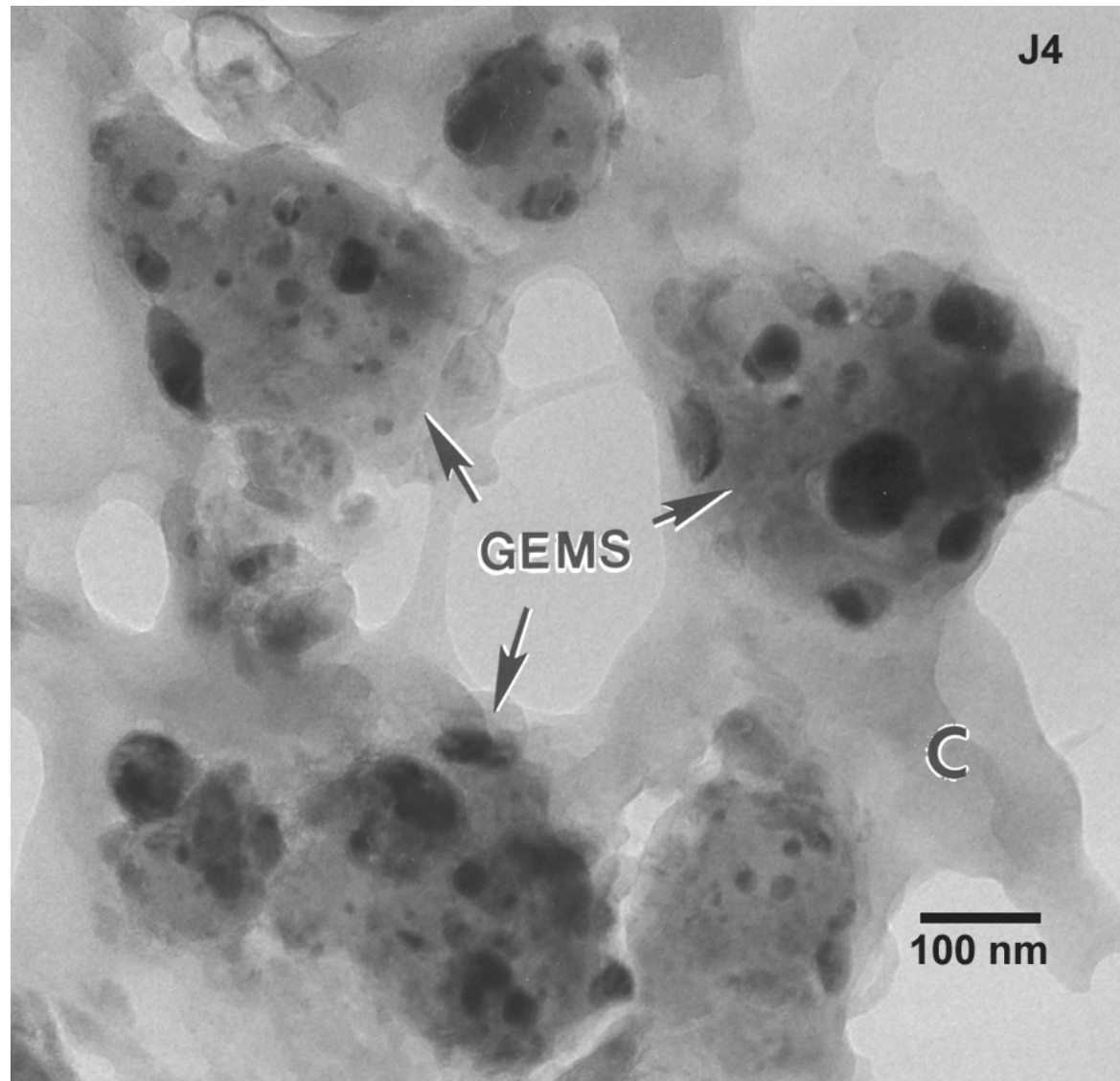


Extinction curves
in the interstellar medium

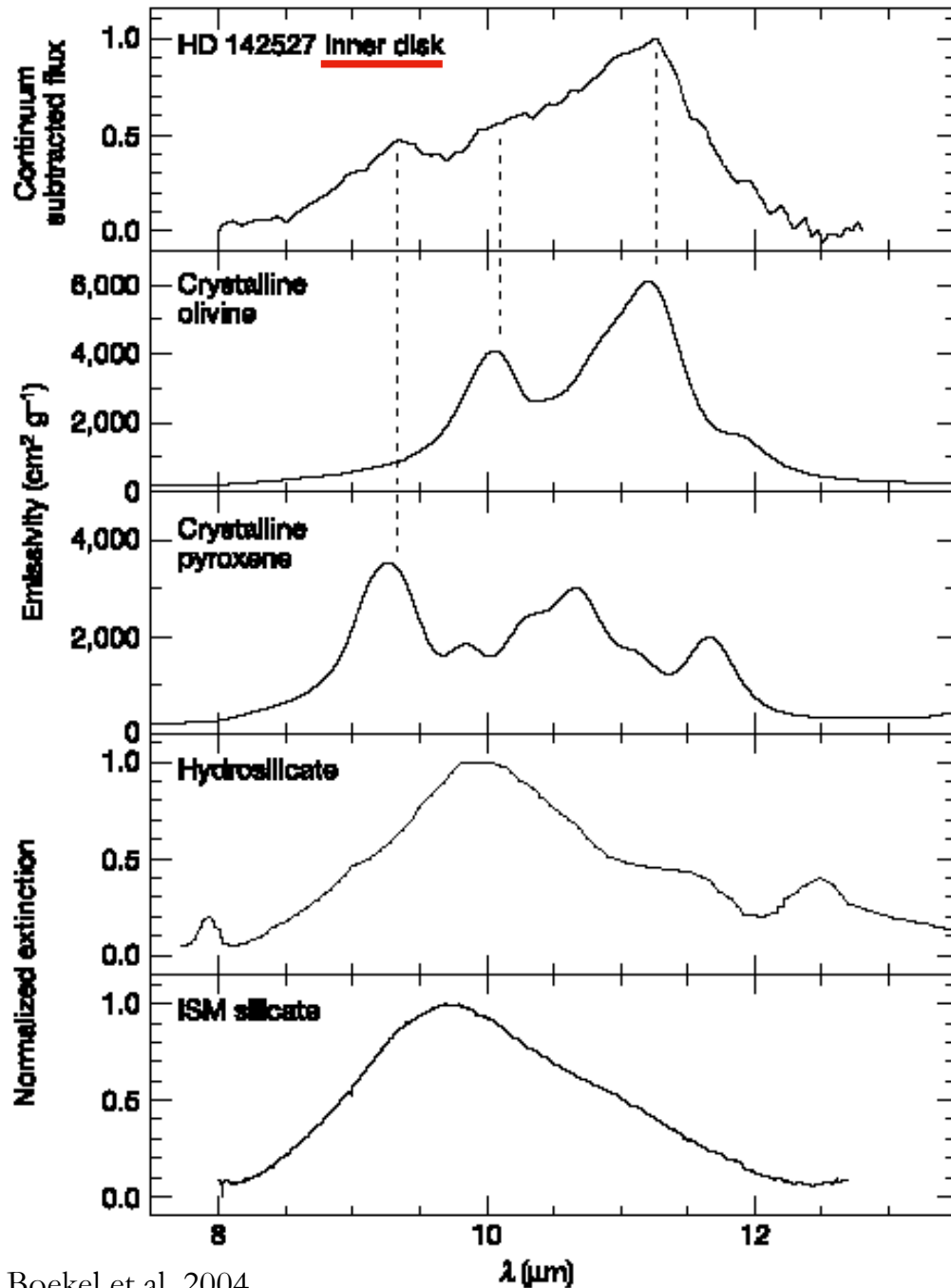
In what form, gas or solids, are the chemical elements in the Solar nebula, i.e. in the parent interstellar molecular cloud?



GEMS (glass with embedded metal sulfides) in IDPs (Interplanetary Dust Particles)



Keller & Messenger, 2005

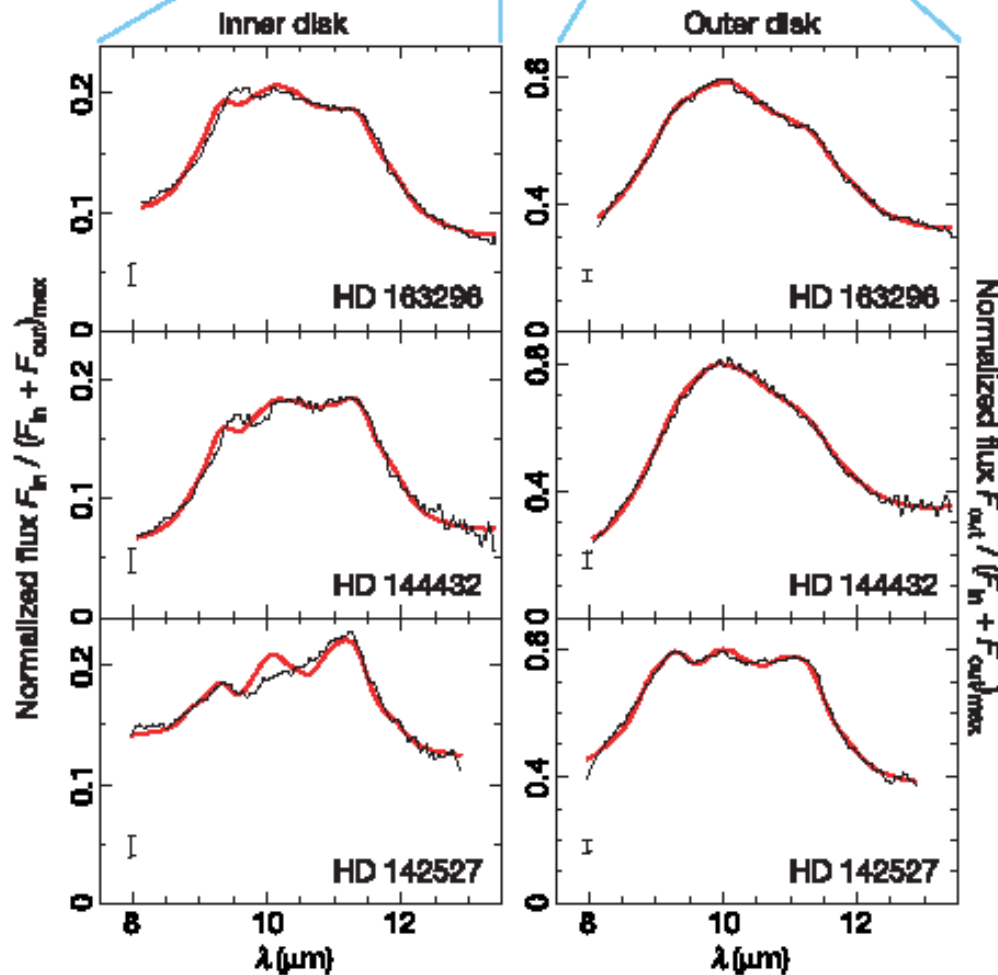
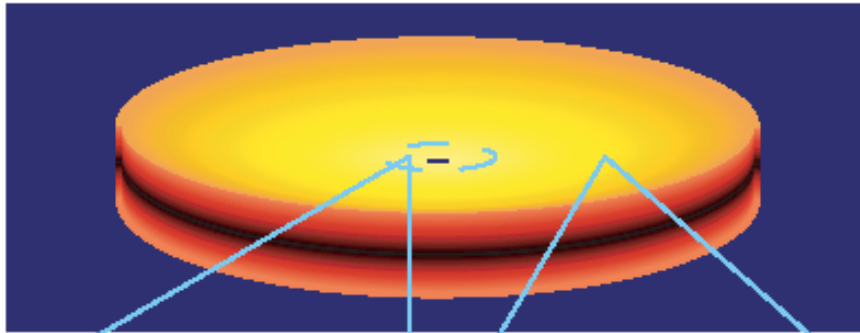


Spectroscopy of the disk of
 (Herbig Ae) HD 142527
 (young star ≈ 1 Ma)
 with VLTI (resolution 1-2 AU)

Main bands for “crystallized“ silicates
 - pyroxenes : $9.3 \mu\text{m}$
 - olivines = $11.2 \mu\text{m}$



1. Crystallized silicates \neq ISM
2. Primary silicates because no band for hydrated silicates (?)

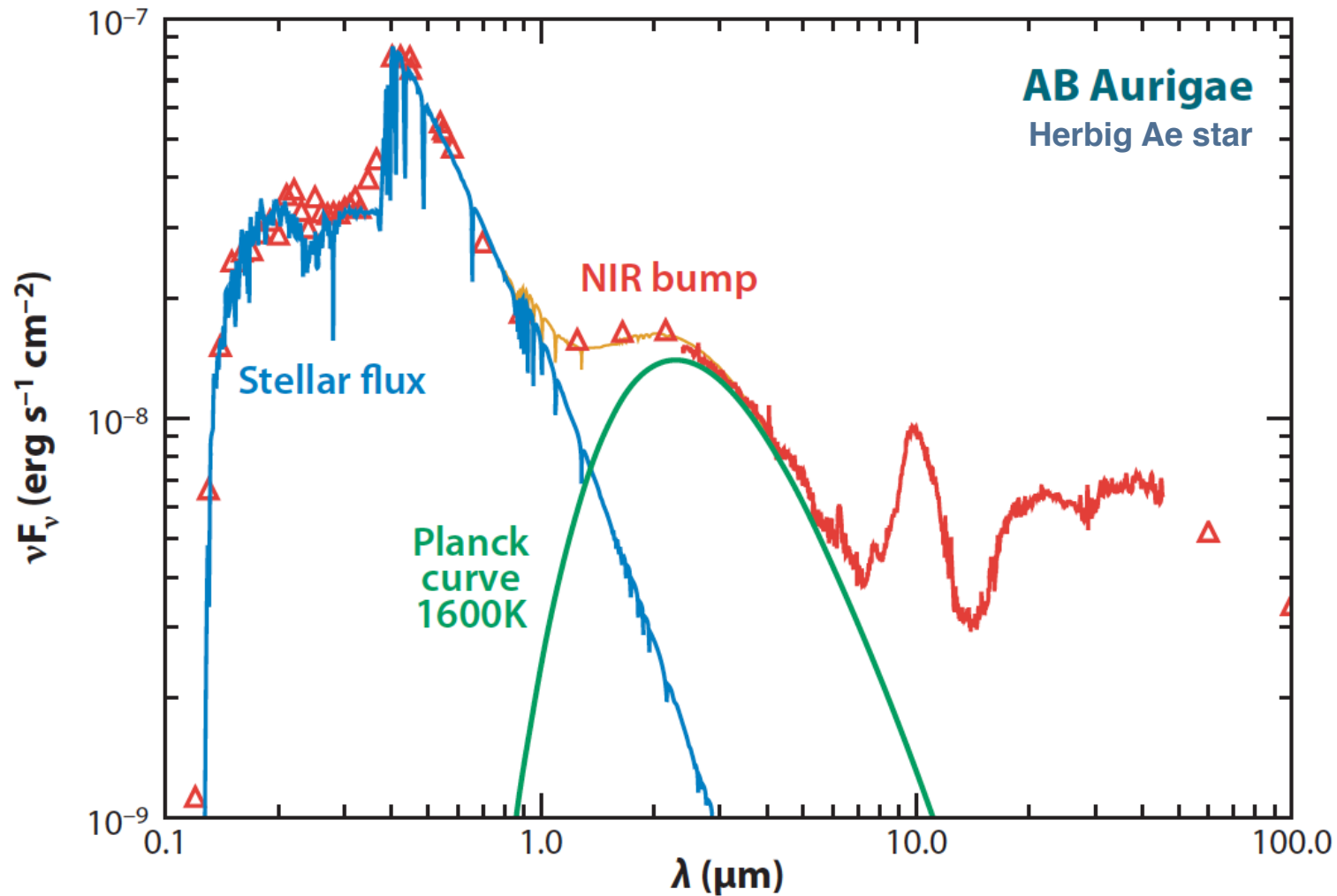


Spectra in the disks of three young (Herbig Ae) stars

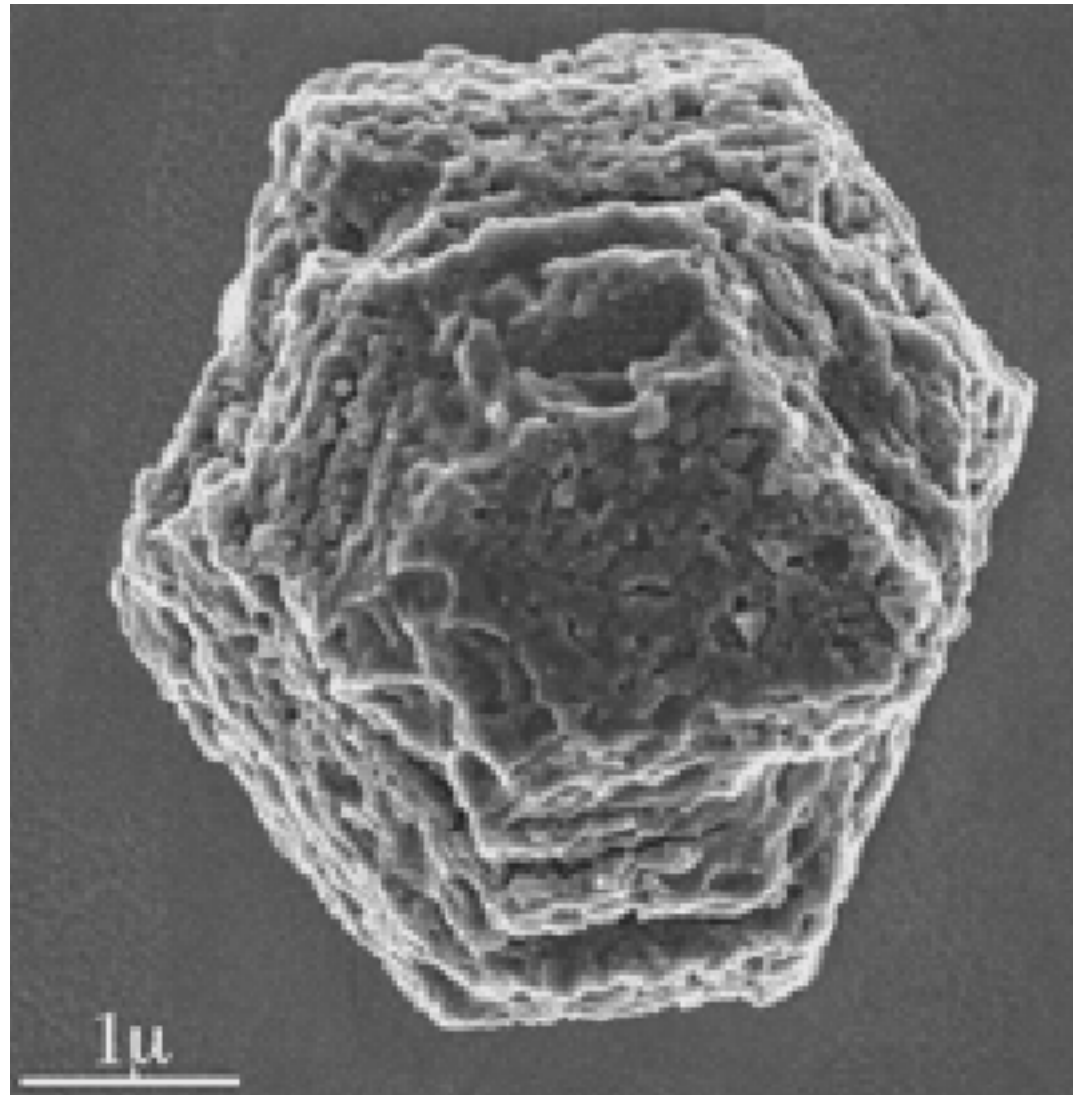


Evolution of amorphous grains in the outer disks to crystallized grains in the inner disks (but over-abundance of crystalline silicates in the outer disk relative to the ISM)

The bump in the IR emission at 1500-1600K is explained by the thermal emission from the dust heated by Sun light (and backwarming). It shows that dust at $T > 1600-1800$ K is missing

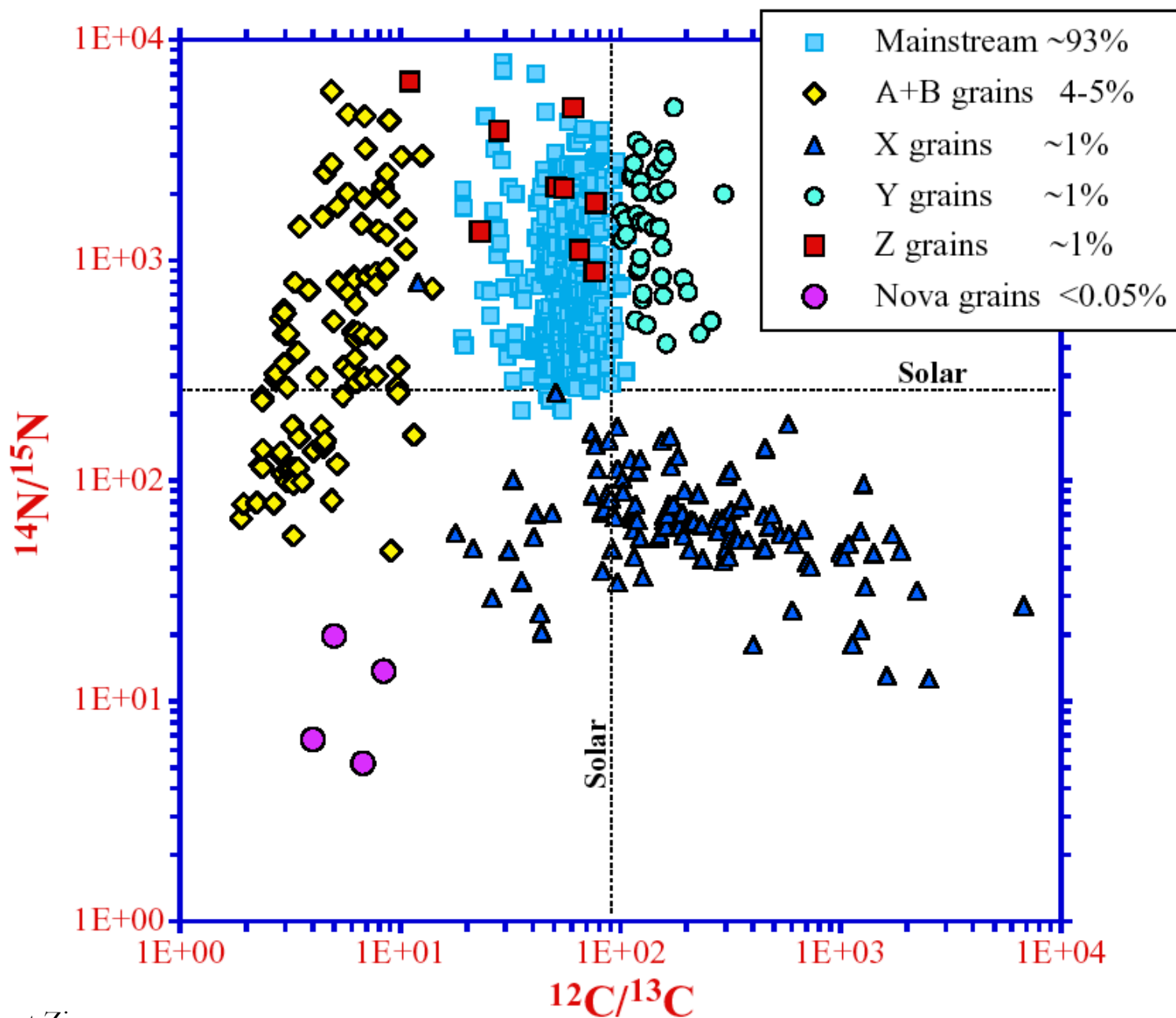


Presolar SiC (after acid treatment)



Bernatovicz et al. (2000)

Presolar SiC grains



Courtesy Ernst Zinner

- Hydrogen fusion ($T = 15 \times 10^6$ K et $d = 150$ g/cm³)

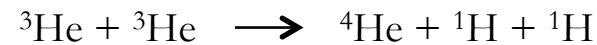
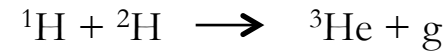
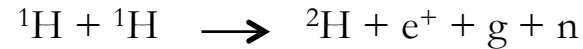


Signature of H fusion

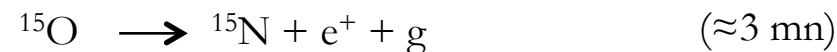
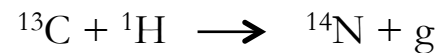
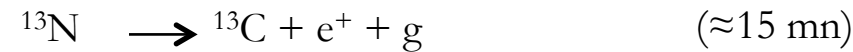
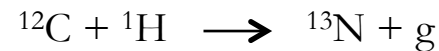
low $^{12}\text{C}/^{13}\text{C}$

high $^{14}\text{N}/^{15}\text{N}$

- P-P cycle ($\approx 85\%$ for $1 M_{\odot}$ stars)



- CNO cycle (dominant for stars of several M_{\odot})



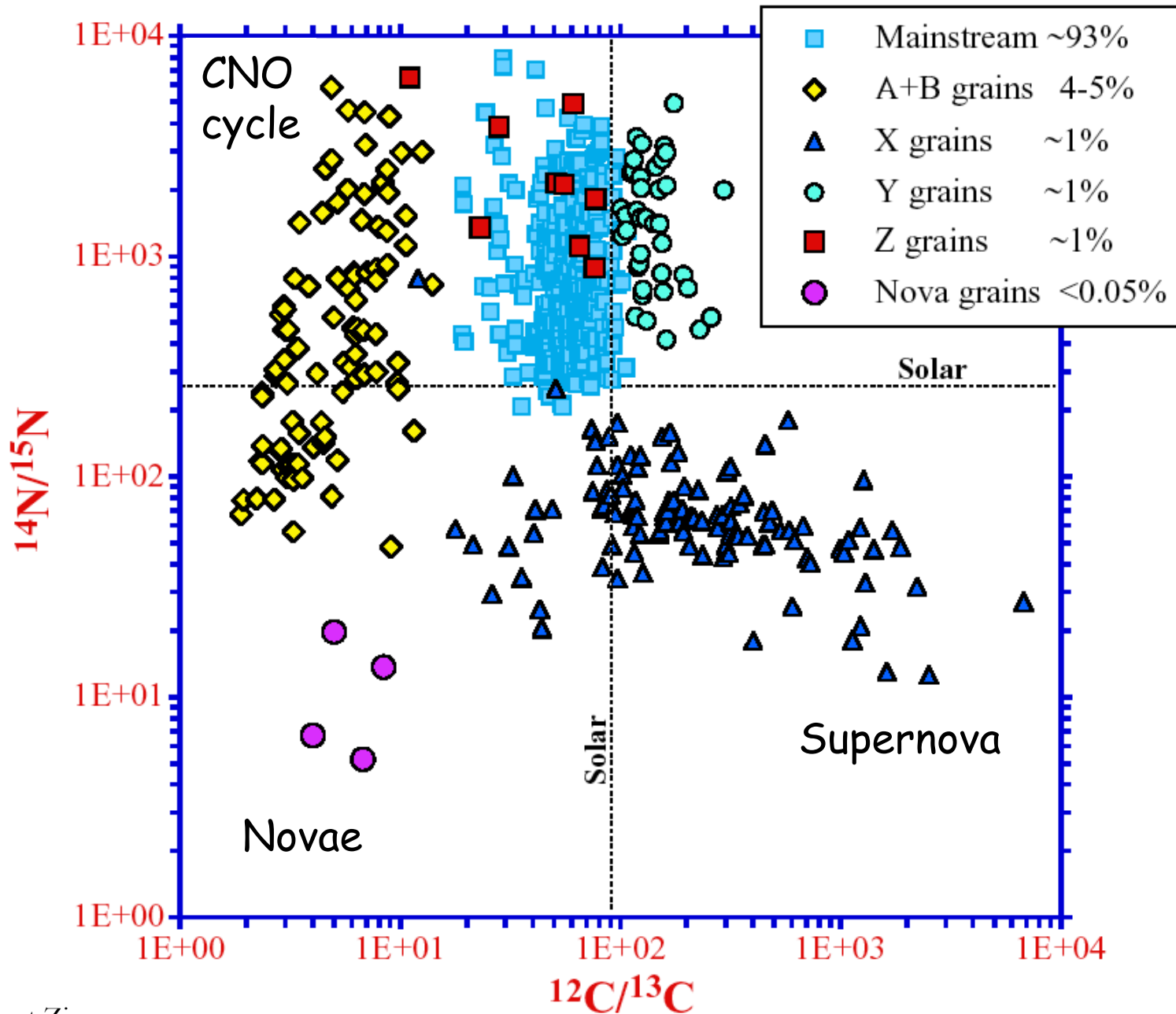
- At equilibrium in the CNO cycle: $^{12}\text{C}/^{13}\text{C} \approx 3.5$

$$^{14}\text{N}/^{15}\text{N} \approx 30\,000 \text{ (low T)} \text{ or } \approx 0.1 \text{ (high T)}$$

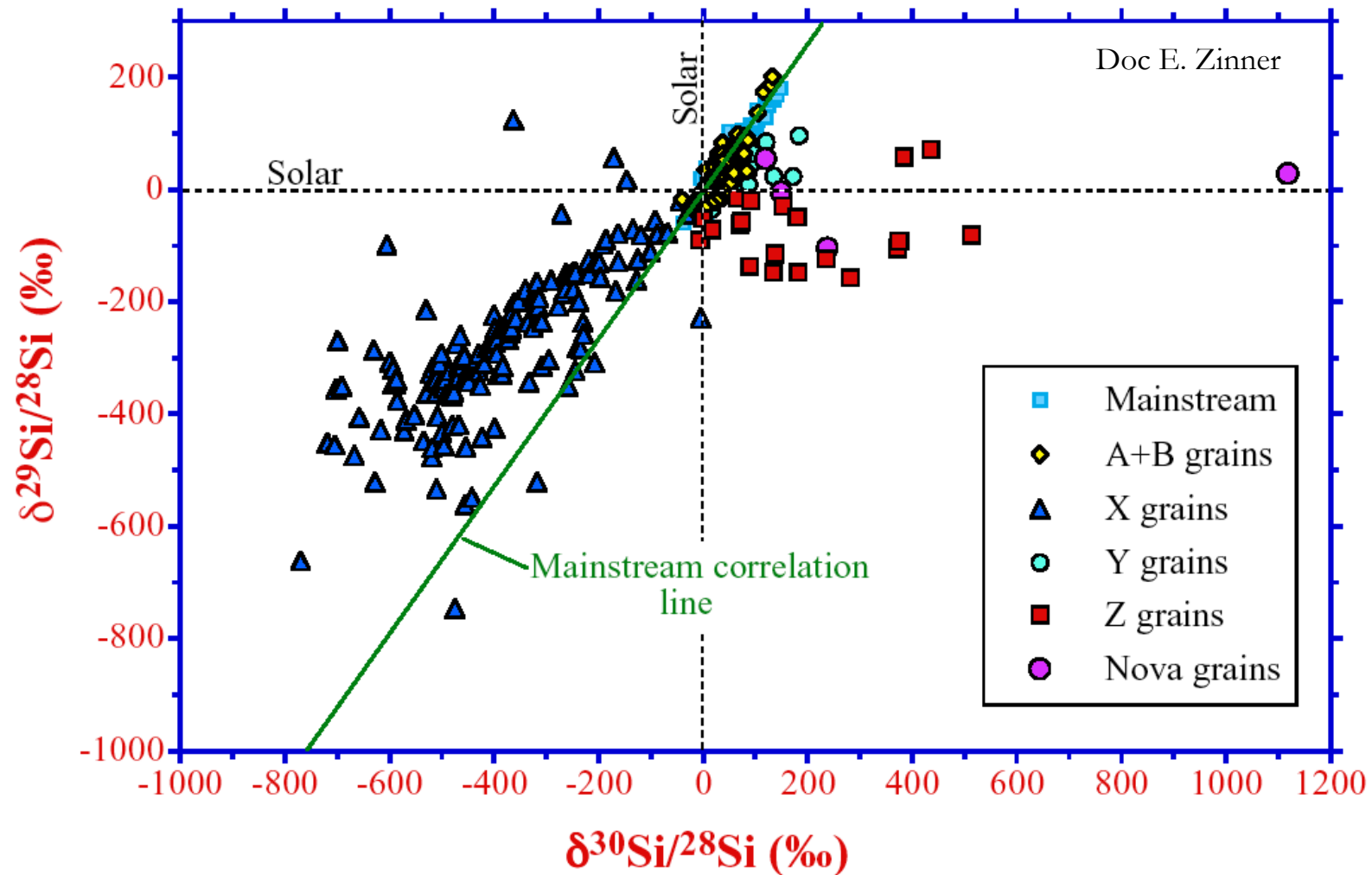
Observed in envelopes of AGB stars: $^{12}\text{C}/^{13}\text{C}$ from ≈ 6 to ≈ 20

$$(^{12}\text{C}/^{13}\text{C})_{\text{Solar}} = 89$$

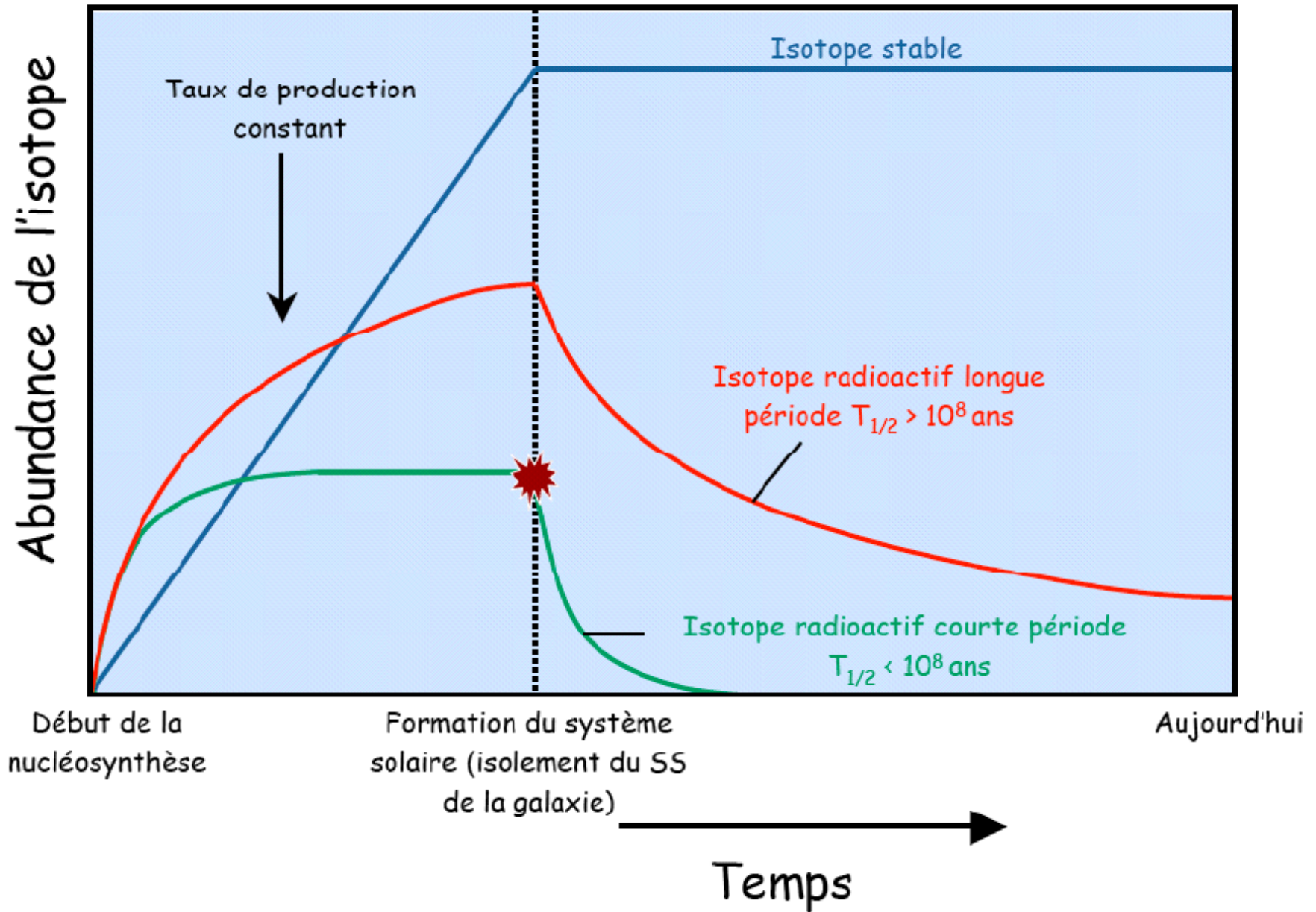
Presolar SiC grains



Presolar SiC grains



- mainstream grains (200‰ variation along a slope ≈ 1.4) while nucleosynthesis in AGB stars makes ^{29}Si and ^{30}Si with a slope $\delta^{29}\text{Si}$ versus $\delta^{30}\text{Si}$ of 0.2-0.5
- grains X : ^{28}Si from supernova



Heavy r-process elements are produced during supernova explosion by neutron capture on ^{56}Fe and subsequent β^- decay of unstable nuclei

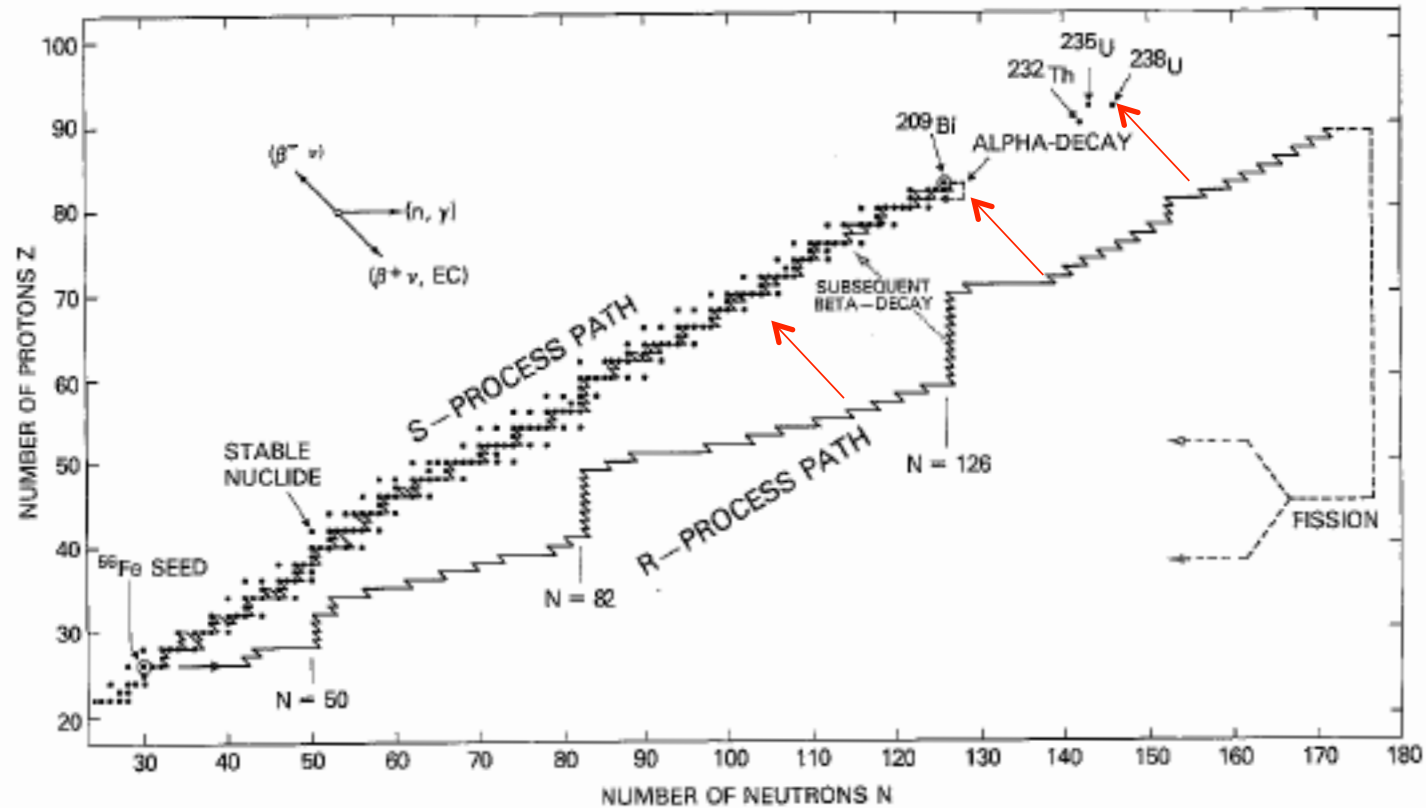
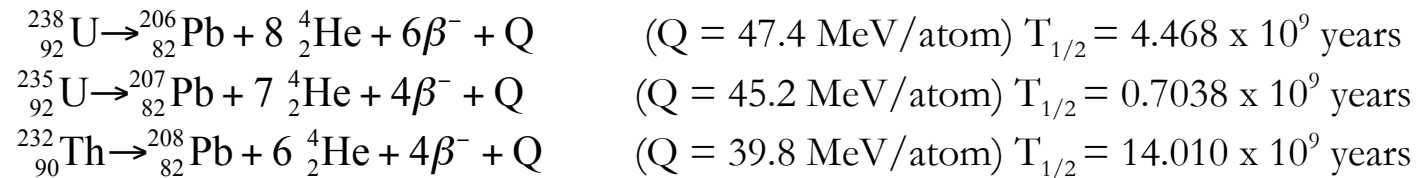


FIGURE 9.13. Neutron-capture paths for the *s*-process and the *r*-process are shown in the (N, Z) -plane. Both paths start with the iron-peak nuclei as seeds (mainly ^{56}Fe). The *s*-process follows a path along the stability line and terminates finally above ^{209}Bi via α -decay (Cla67). The *r*-process drives the nuclear matter far to the neutron-rich side of the stability line, and the neutron capture flows upward in the (N, Z) -plane until β -delayed fission and neutron-induced fission occur (Thi83). The *r*-process path shown was computed (See65) for the conditions $T_9 = 1.0$ and $N_n = 10^{24}$ neutrons cm^{-3} .

Ratios of heavy r-process elements U & Th can be used to date the galaxy because (i) their half-lives are long enough, (ii) their production rates can be determined quite precisely from theoretical models and observations and can be considered constant, (iii) the time during which they are produced can be considered negligible relative to the age of the galaxy, (iv) their ratios in meteorites are resistant to secondary perturbations.



- Today in chondrites: ${}^{238}\text{U} / {}^{232}\text{Th} = 0.270$ and ${}^{235}\text{U} / {}^{238}\text{U} = 0.007253$
- From now back to 4.567 Gy ago: radioactive decay

$$\begin{aligned}
 \left(\frac{{}^{238}\text{U}}{{}^{232}\text{Th}} \right)_{4.57} &= \left(\frac{{}^{238}\text{U}}{{}^{232}\text{Th}} \right)_0 \times \exp^{(\lambda_{238} - \lambda_{232}) \times 4.57 \times 10^9} = 0.438 \\
 \left(\frac{{}^{235}\text{U}}{{}^{238}\text{U}} \right)_{4.57} &= \left(\frac{{}^{235}\text{U}}{{}^{238}\text{U}} \right)_0 \times \exp^{(\lambda_{235} - \lambda_{238}) \times 4.57 \times 10^9} = 0.3216
 \end{aligned}$$

- From 4.567 Gy ago to the birth of the galaxy: radioactive decay + production in SN

$$\frac{d[Z]}{dt} = y\omega - \lambda[Z]$$

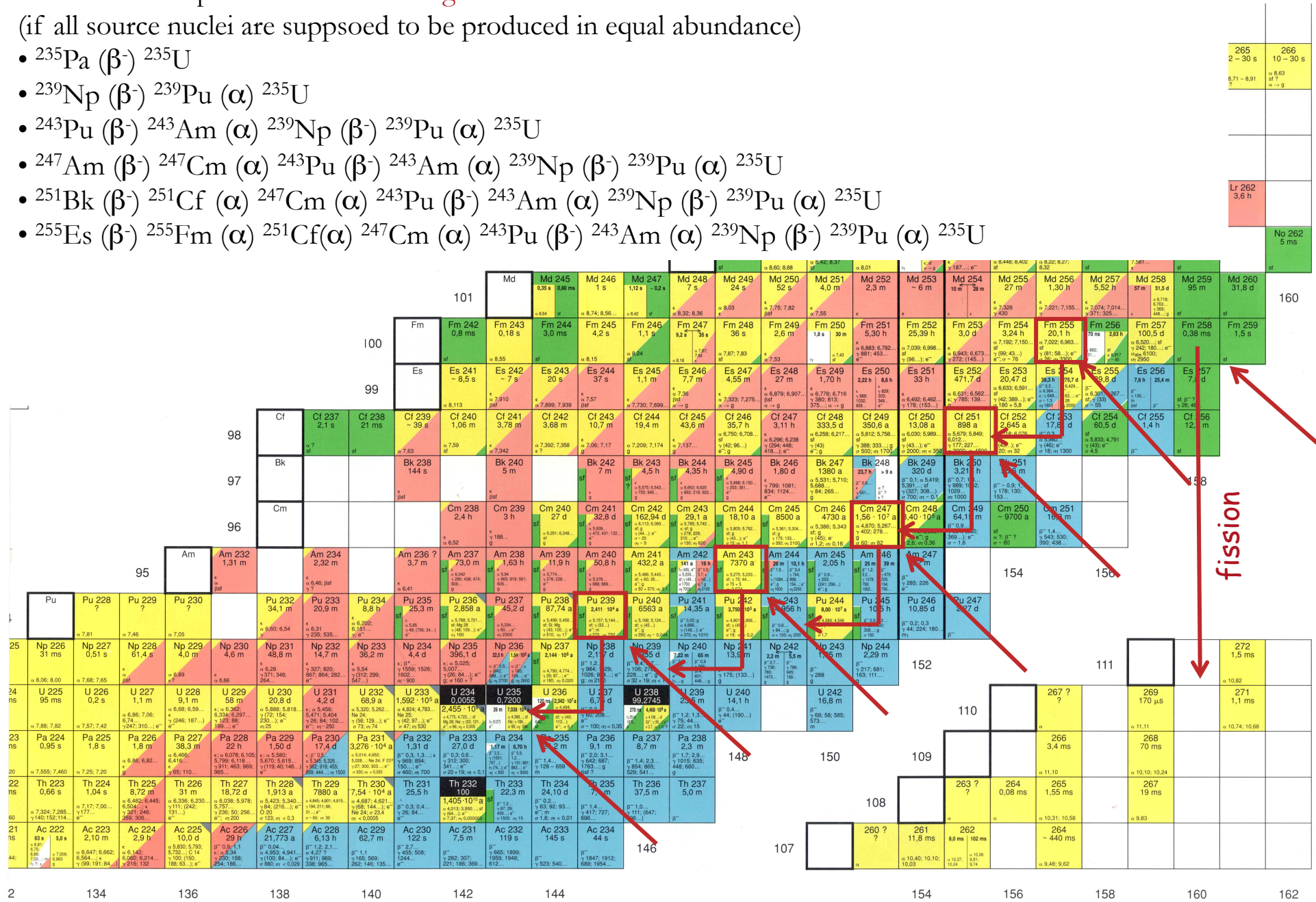
y = nucleosynthetic yield for isotope Z

ω = rate of star formation

$$[Z] = \frac{y\omega}{\lambda} \left(1 - \exp^{-\lambda t} \right)$$

One event of r-process makes 6 "Progenitors" of ^{235}U
 (if all source nuclei are supposed to be produced in equal abundance)

- ^{235}Pa (β^-) ^{235}U
- ^{239}Np (β^-) ^{239}Pu (α) ^{235}U
- ^{243}Pu (β^-) ^{243}Am (α) ^{239}Np (β^-) ^{239}Pu (α) ^{235}U
- ^{247}Am (β^-) ^{247}Cm (α) ^{243}Pu (β^-) ^{243}Am (α) ^{239}Np (β^-) ^{239}Pu (α) ^{235}U
- ^{251}Bk (β^-) ^{251}Cf (α) ^{247}Cm (α) ^{243}Pu (β^-) ^{243}Am (α) ^{239}Np (β^-) ^{239}Pu (α) ^{235}U
- ^{255}Es (β^-) ^{255}Fm (α) ^{251}Cf (α) ^{247}Cm (α) ^{243}Pu (β^-) ^{243}Am (α) ^{239}Np (β^-) ^{239}Pu (α) ^{235}U



265
2-30 s
8.71-8.91

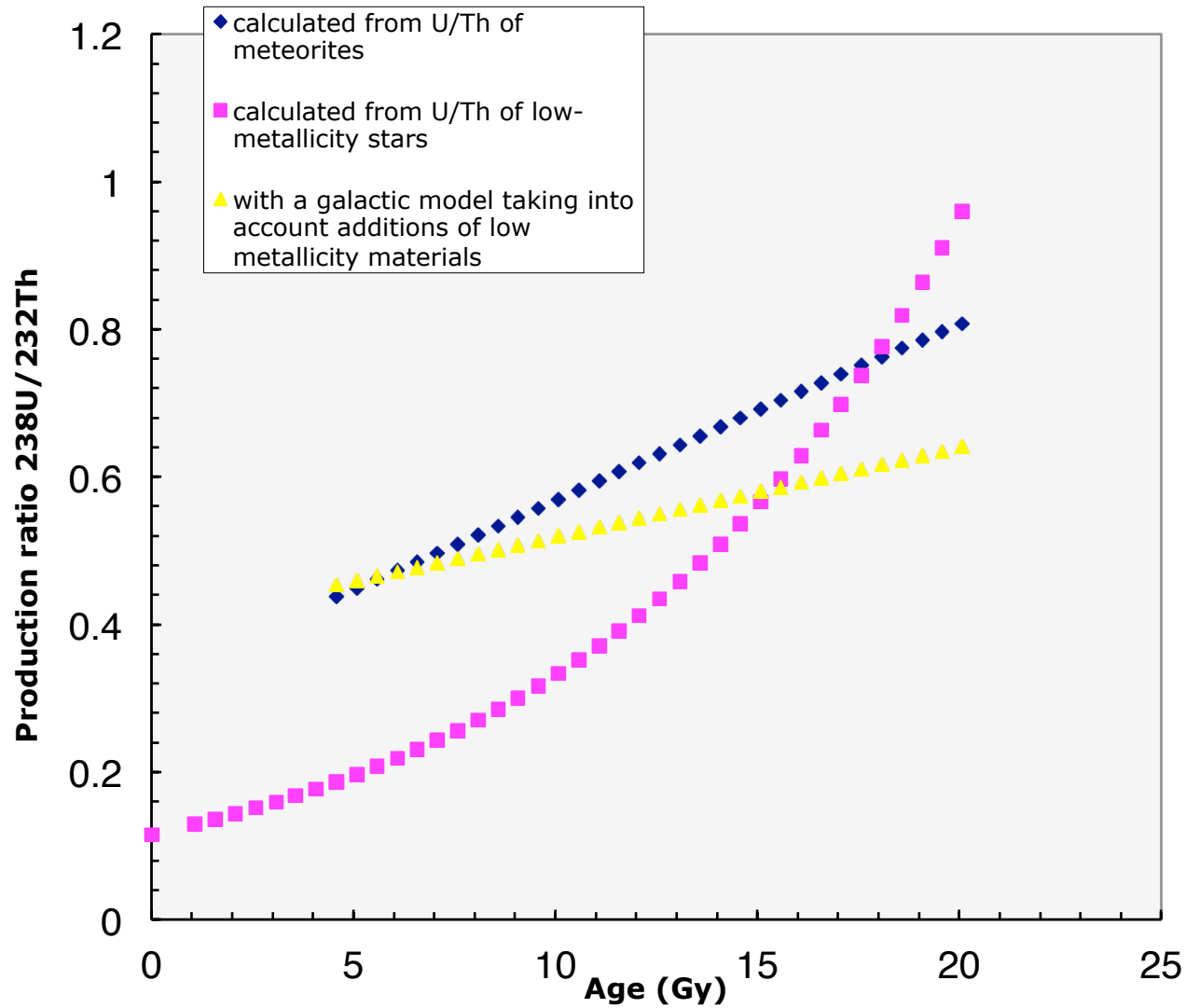
266
10-30 s
8.63
st
α-g

Lr 262
3.6 h

No 262
5 ms

fission

$$P(^{235}\text{U})/P(^{238}\text{U}) = 6/3.1 = 1,9 \text{ (instead of theoretical value of 1.506)}$$
$$P(^{238}\text{U})/P(^{232}\text{Th}) = 3.1/5 \approx 0.6 \text{ (instead of theoretical value of 0.578)}$$



How is the condensation working?

The condensation sequence (Larimer 1967, Grossman 1972)

- two hypotheses:

- $P_{H_2} \approx P_{total} \approx 10^{-3} - 10^{-5} P_{at}$ (H is 1000 times more abundant than all other elements, except He which is not considered, H_2 is more abundant than all other H-bearing species)
- effects des P, T, X gradients and turbulence ignored

- use of the ideal gas law (valid at these low pressures)

- for a given volume:

$$N_{H_2} = P_{H_2} / RT \text{ and } N_H^{tot} = 2 N_{H_2}$$

- for any element X:

$$N_X^{tot} = [X] / [H] \times N_H^{tot}$$

- for any element, mass balance gives:

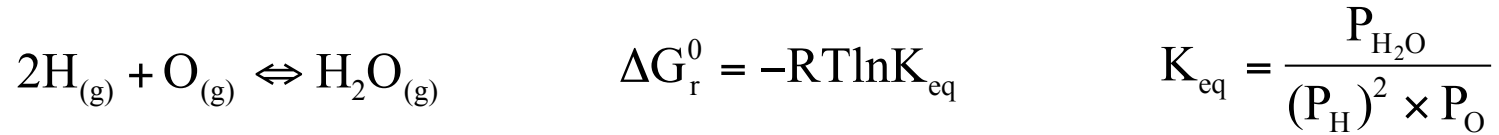
$$\text{e.g. for O: } N_O^{tot} = N_O + N_{H_2O} + N_{CO} + 2N_{CO_2} + N_{org} + \dots$$

Table 1. Gaseous species contributing more than 10^{-7} of the total moles of their common constituent element between 2000°K and 1200°K .

Element	Abundance* (Si = 10^6)	Gaseous species
Hydrogen	2.6×10^{10}	$\text{H}_2, \text{H}, \text{H}_2\text{O}, \text{HF}, \text{HCl}, \text{MgH}, \text{HS}, \text{H}_2\text{S}, \text{MgOH}$
Oxygen	2.36×10^7	$\text{CO}, \text{SiO}, \text{H}_2\text{O}, \text{TiO}, \text{OH}, \text{HCO}, \text{CO}_2, \text{PO}, \text{CaO}, \text{COS}, \text{MgO}, \text{SiO}_2, \text{AlOH}, \text{SO}$ $\text{NaOH}, \text{MgOH}, \text{PO}_2, \text{Mg}(\text{OH})_2, \text{AlO}_2\text{H}$
Carbon	1.35×10^7	$\text{CO}, \text{HCN}, \text{CS}, \text{HCO}, \text{CO}_2, \text{COS}, \text{HCP}$
Nitrogen	2.44×10^6	$\text{N}_2, \text{HCN}, \text{PN}, \text{NH}_3, \text{NH}_2$
Magnesium	1.05×10^6	$\text{Mg}, \text{MgH}, \text{MgS}, \text{MgF}, \text{MgCl}, \text{MgO}, \text{MgOH}, \text{Mg}(\text{OH})_2$
Silicon	1.00×10^6	$\text{Si}, \text{SiS}, \text{SiO}, \text{SiO}_2$
Iron	8.90×10^5	Fe
Sulfur	5.06×10^5	$\text{SiS}, \text{CS}, \text{S}, \text{HS}, \text{H}_2\text{S}, \text{PS}, \text{AlS}, \text{MgS}, \text{NS}, \text{S}_2, \text{COS}, \text{SO}, \text{CS}_2, \text{SO}_2$
Aluminum	8.51×10^4	$\text{Al}, \text{AlH}, \text{AlF}, \text{AlCl}, \text{AlS}, \text{AlO}, \text{Al}_2\text{O}, \text{AlOH}, \text{AlOF}, \text{AlO}_2\text{H}$
Calcium	7.36×10^4	$\text{Ca}, \text{CaF}, \text{CaO}, \text{CaCl}_2$
Sodium	6.32×10^4	$\text{Na}, \text{NaH}, \text{NaCl}, \text{NaF}, \text{NaOH}$
Nickel	4.57×10^4	Ni
Phosphorus	1.27×10^4	$\text{P}, \text{PN}, \text{PH}, \text{P}_2, \text{PH}_2, \text{PS}, \text{PO}, \text{PH}_3, \text{PO}_2, \text{HCP}$
Chromium	1.24×10^4	Cr
Manganese	8800	Mn
Fluorine	3630	$\text{HF}, \text{AlF}, \text{CaF}, \text{F}, \text{MgF}, \text{NaF}, \text{NF}, \text{KF}, \text{PF}, \text{CaF}_2, \text{AlOF}, \text{TiF}_2, \text{MgF}_2,$ MgClF, TiF
Potassium	3240	$\text{K}, \text{KH}, \text{KCl}, \text{KF}, \text{KOH}$
Titanium	2300	$\text{Ti}, \text{TiO}, \text{TiF}_2, \text{TiO}_2, \text{TiF}$
Cobalt	2300	Co
Chlorine	1970	$\text{HCl}, \text{Cl}, \text{AlCl}, \text{NaCl}, \text{KCl}, \text{MgCl}, \text{CaCl}_2, \text{MgCl}_2, \text{AlOCl}, \text{MgClF}$

* From CAMERON (1968).

- All reactions are considered to happen at equilibrium with mono-atomic gaseous species for instance for water:

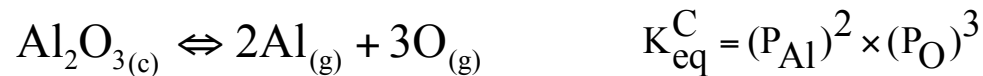


so that: $N_{\text{H}_2\text{O}} = K_{\text{eq}} (N_{\text{H}})^2 N_{\text{O}} (RT)^2$

- The abundances of all gaseous species can be calculated in function of T for a given P and bulk composition.

- Then the solids that are stable for these P_x and T are calculated

For instance for corundum:



K_{eq}^{C} is calculated from the composition of the gas: $\ln K_{\text{eq}}^{\text{C}} = 2 \ln(P_{\text{Al}}) + 3 \ln(P_{\text{O}})$

K_{eq}^{C} is compared with the value of K_{eq} calculated from free energy: $\ln K_{\text{eq}} \approx A + \frac{B}{T}$

If $K_{\text{eq}}^{\text{C}} > K_{\text{eq}}$ \Rightarrow condensation \Rightarrow T is determined

- The calculation continues assuming constant thermodynamic equilibrium with the gas

At 1700 K, $(P_{\text{Al}})^{1700\text{K}} < (P_{\text{Al}})^{1758\text{K}}$, a fraction of Al has been removed from the gas: mass balance must be written

$$(N_{\text{Al}_2\text{O}_3})^{1700\text{K}} = \frac{1}{2}(N_{\text{Al}})^{1758\text{K}} - \frac{1}{2}(N_{\text{Al}})^{1700\text{K}} = \frac{1}{2R} \left(\frac{(P_{\text{Al}})^{1758\text{K}}}{1758} - \frac{(P_{\text{Al}})^{1700\text{K}}}{1700} \right)$$

- The ‘classical’ 50% temperature condensation (T_{50}) is the temperature for which 50% of the element is condensed.

This is not a property of the element but of the minerals in which the element is scavenged during condensation (which depends on bulk composition of the gas)

MAJOR ELEMENT CONDENSATION TEMPERATURES

Ideal Formula	Mineral Name	Solar System Composition (K)	Photospheric Composition (K)
Al ₂ O ₃	Corundum	1677	1665
CaAl ₁₂ O ₁₉	Hibonite	1659	1647
CaAl ₄ O ₇	Grossite	1542	1531
Ca ₂ Al ₂ SiO ₇	Gehlenite	1529	1519
CaTiO ₃	Perovskite	1593	1584
Ca ₄ Ti ₃ O ₁₀	Ca titanate	1578	1567
Ca ₃ Ti ₂ O ₇	Ca titanate	1539	1529
Ca ₄ Ti ₃ O ₁₀	Ca titanate	1512	1502
CaTiO ₃	Perovskite	1441	1429
MgAl ₂ O ₄	Spinel	1397	1387
CaAl ₂ Si ₂ O ₈	Anorthite	1387	1378
Mg ₂ SiO ₄	Forsterite	1354	1346
MgSiO ₃	Enstatite	1316	1308
CaMgSi ₂ O ₆	Diopside	1347	1339
Fe	Fe alloy	1357	1351
Fe ₃ P	Schreibersite	1248	1245
FeS	Troilite	704	693
Fe ₃ O ₄	Magnetite	371	365
H ₂ O	Water ice	182	181

NOTE.—At 10⁻⁴ bar total pressure.

For most elements, the interval of condensation is small ($\approx 10\text{-}50\text{ K}$)

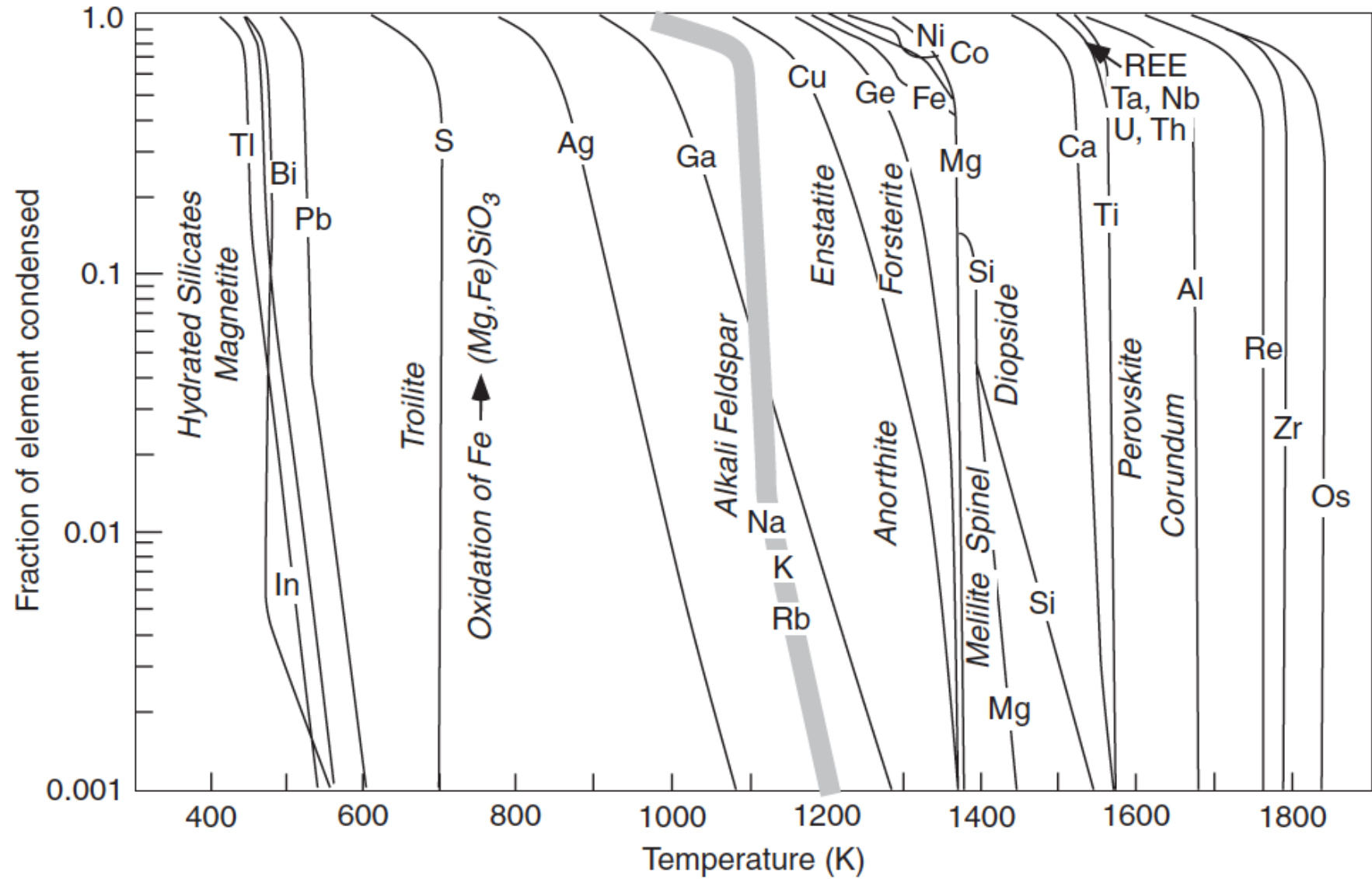
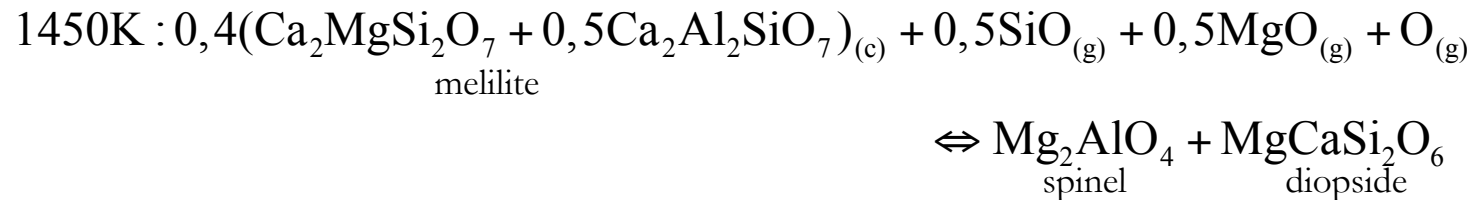


Fig from McSween & Huss 2010 modified from Grossman & Larimer 1974

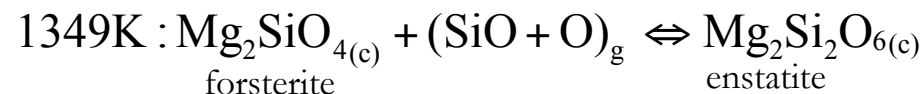
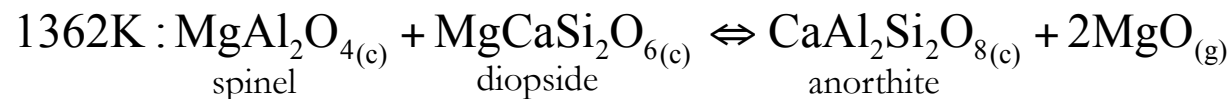
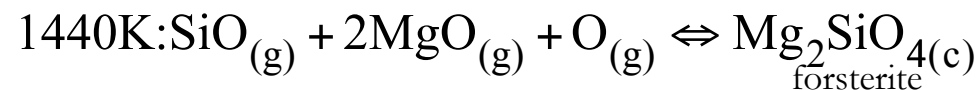
- a “phase diagram” of the Solar nebula is built by calculating which minerals is stable for a given P-T-X paths. There are strong effects of composition, pressure (condensing solids or liquids). The phase rule is used in this diagram to predict the minerals at equilibrium.

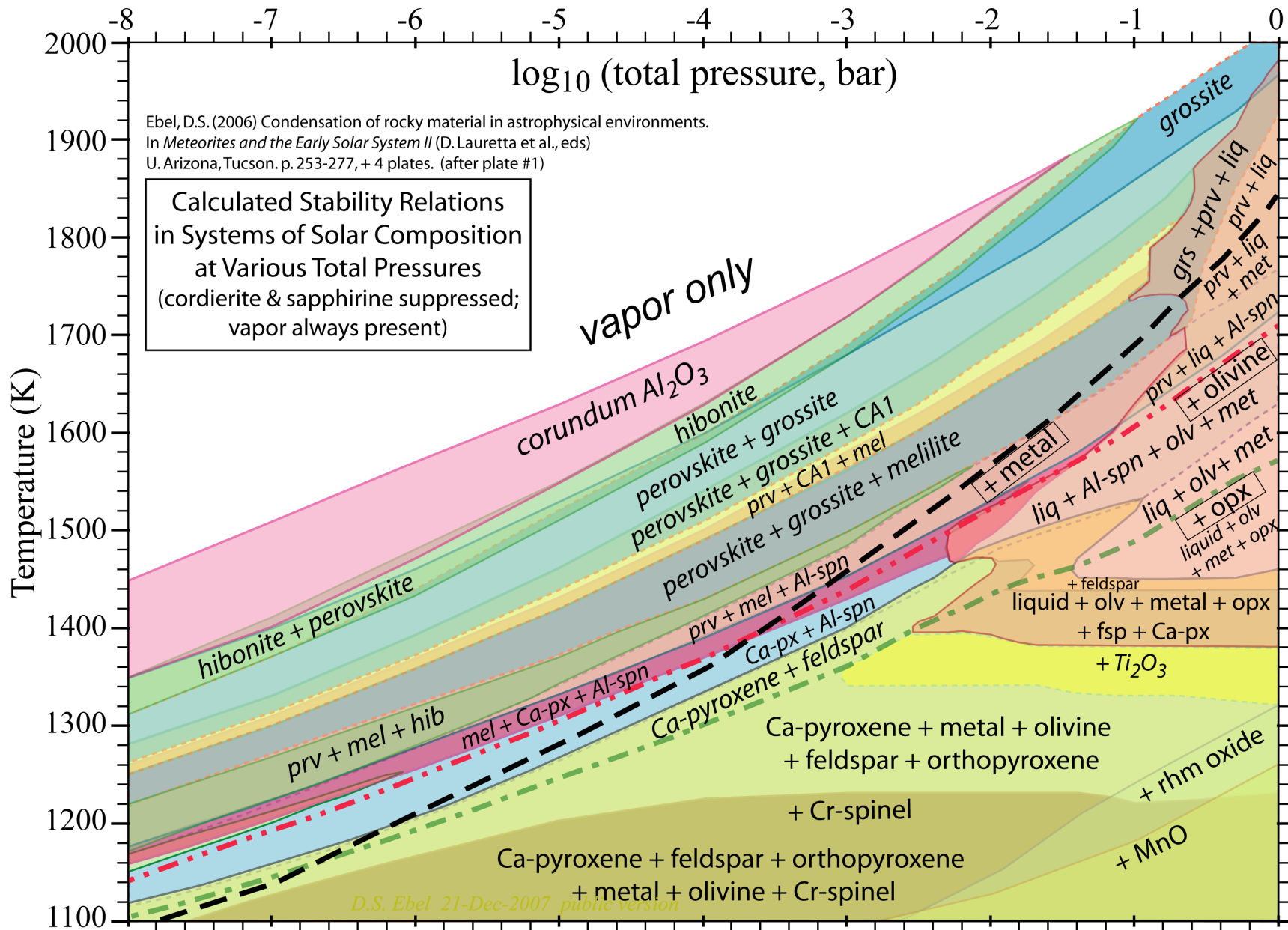


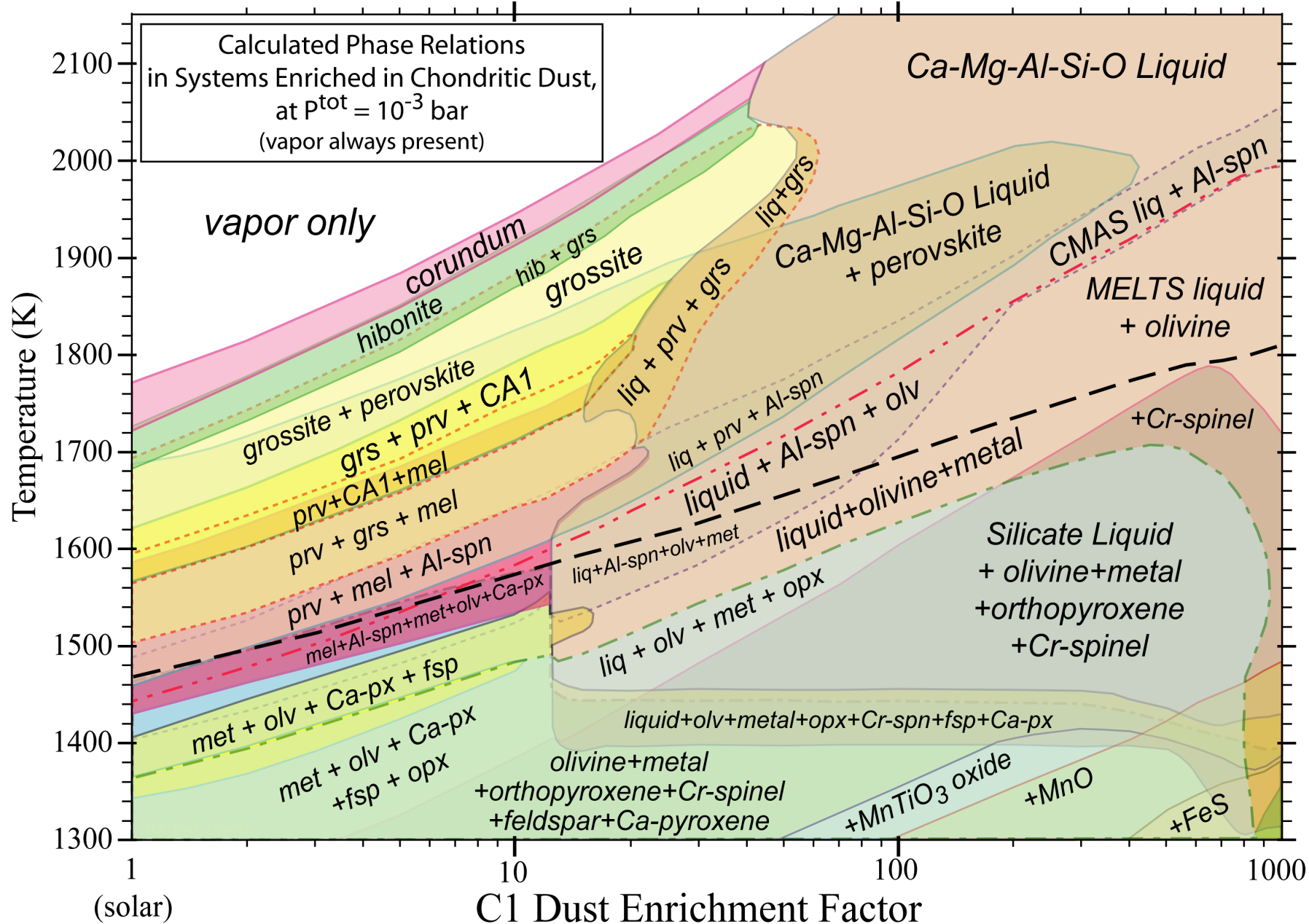
The first Mg-bearing minerals condensing are melilite and spinel (but they contain only a few % of total Mg. Melilite reacts away at 1450K.

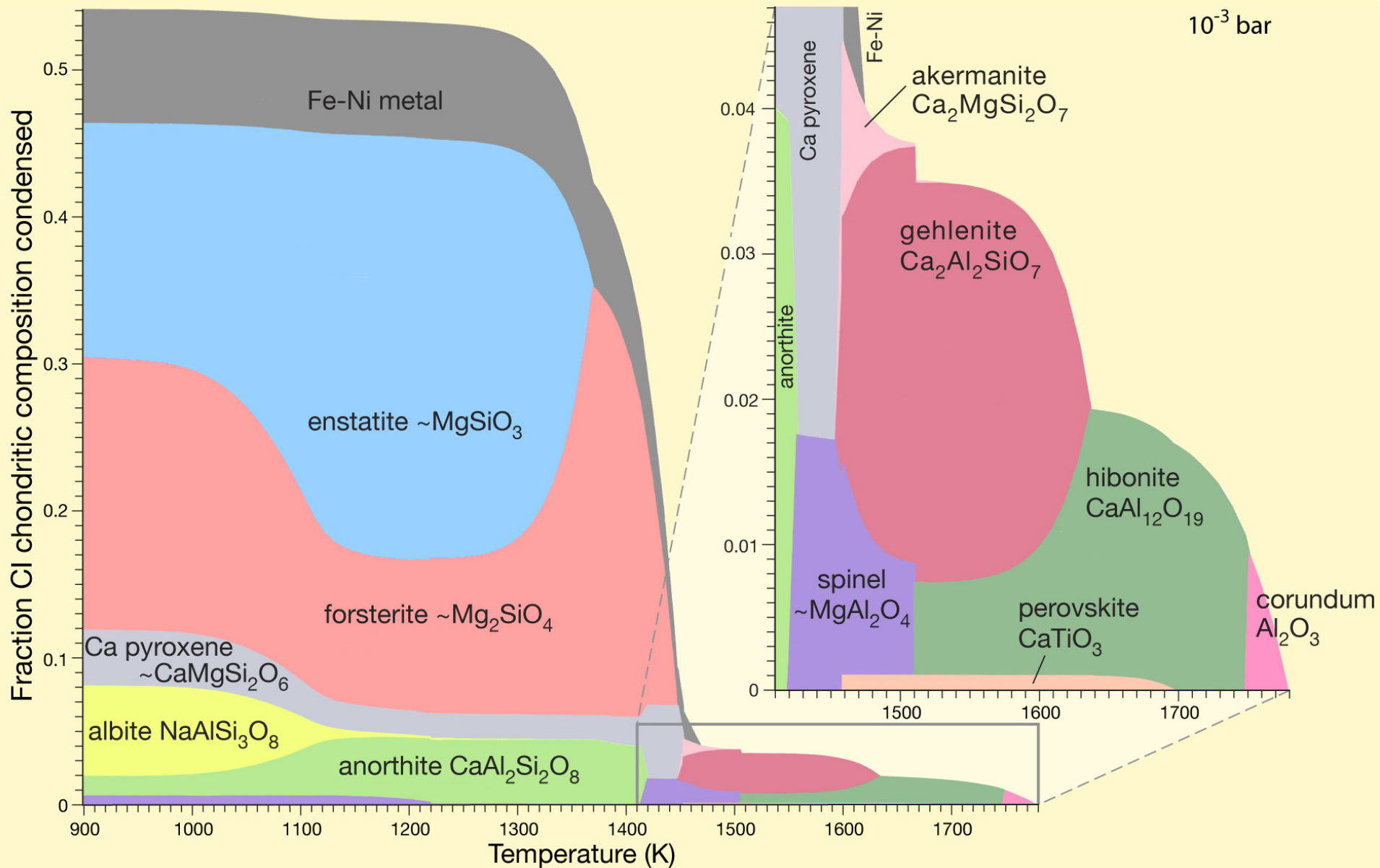


Forsterite appears rapidly at 1440K and scavenges rapidly all Mg from the gas. At 1362K spinel and diopside react and liberate Mg to the gas, Mg which is used to produce more forsterite. At 1349 K forsterite starts to react away with the gas to form enstatite, ...





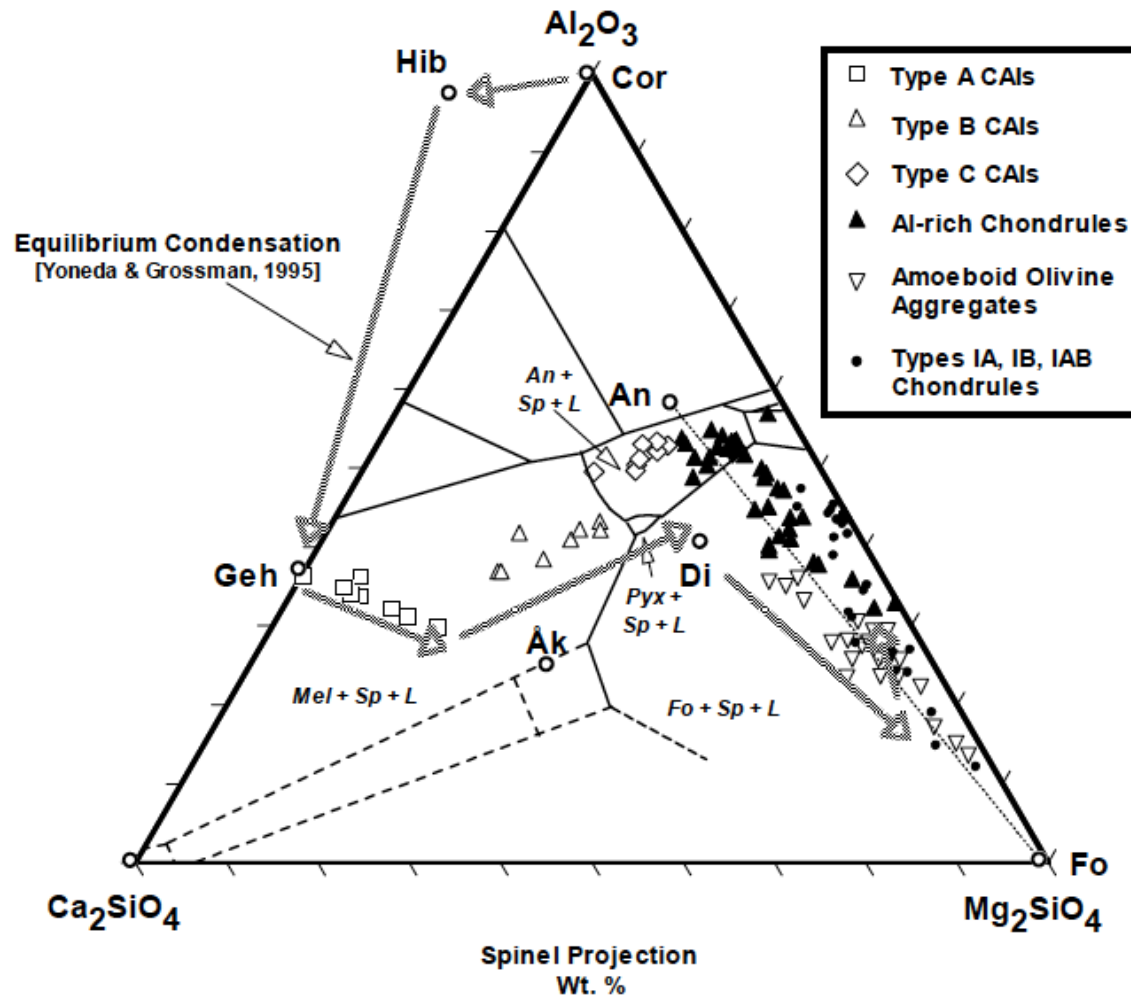




(Davis & Richter, 2003)

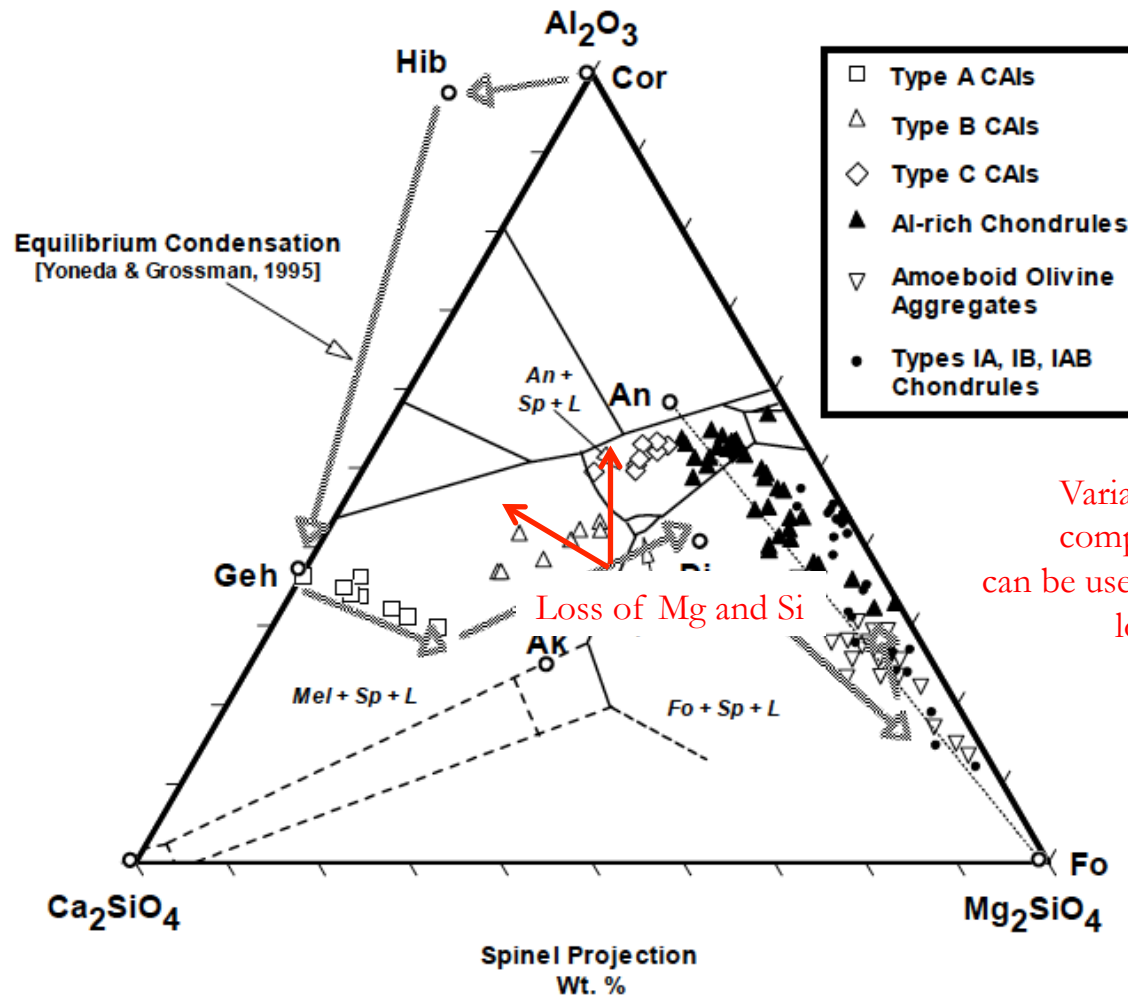
The problems with the condensation sequence

- CAIs do exist: part of condensation happened out of equilibrium
- the “cosmochemical trend” in chondrites suggest fractional condensation
- the composition of CAIs and other high temperature condensates (e.g. AOAs: Amoeboid Olivine Aggregates) differs from predictions made from the condensation sequence



The problems with the condensation sequence

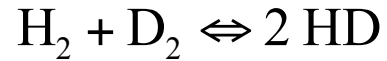
- CAIs do exist: part of condensation happened out of equilibrium
- the composition of CAIs and other high temperature condensates (e.g. AOAs: Ameboid Olivine Aggregates) differs from predictions made from the condensation sequence



Variations of the isotopic compositions of Mg & Si can be used to trace the evaporation loss of Mg and Si

What is isotopic fractionation?

The simplest example

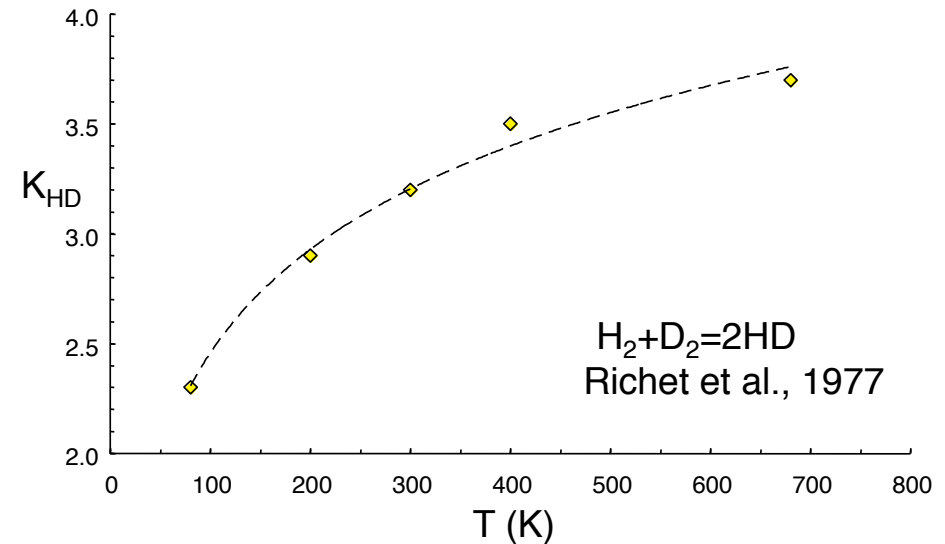


$$P(\text{H}_2) = \frac{N_{\text{H}} \times (N_{\text{H}} - 1)}{N_{\text{T}} \times (N_{\text{T}} - 1)} \approx \frac{N_{\text{H}}^2}{N_{\text{T}}^2}$$

$$P(\text{D}_2) = \frac{N_{\text{D}} \times (N_{\text{D}} - 1)}{N_{\text{T}} \times (N_{\text{T}} - 1)} \approx \frac{N_{\text{D}}^2}{N_{\text{T}}^2}$$

$$P(\text{HD}) = \frac{2 \times N_{\text{D}} \times N_{\text{H}}}{N_{\text{T}} \times (N_{\text{T}} - 1)} \approx \frac{2 \times N_{\text{D}} \times N_{\text{H}}}{N_{\text{T}}^2}$$

$$K_{\text{HD}} = \frac{[\text{HD}]^2}{[\text{H}_2] \times [\text{D}_2]} = \frac{P(\text{HD})^2}{P(\text{H}_2) \times P(\text{D}_2)} = 4$$

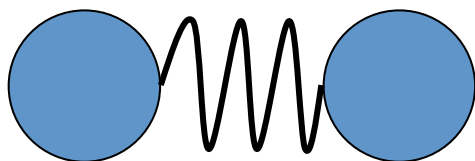


There is more D_2 than predicted from random distribution of H & D isotopes

This is due to the higher stability of the D_2 molecule

The H bond is vibrating: the most stable situation is when the electronic cloud of the covalent bond is in between the two atoms

$$F = -kx$$



Similar at low T to the harmonic oscillator
(except that the frequencies of vibration are quantified)

$$\mu = \frac{m_1 \times m_2}{m_1 + m_2} \quad \nu = \frac{1}{2\pi} \times \sqrt{\frac{k}{\mu}}$$

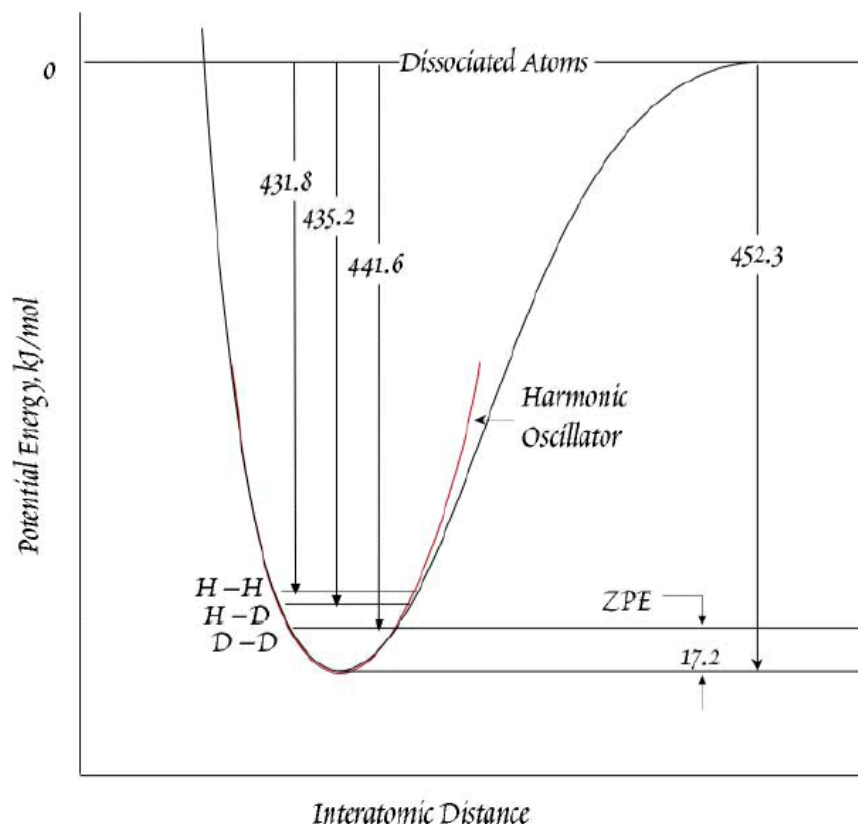
$$\mu_{\text{H}_2} \approx 1/2, \mu_{\text{HD}} \approx 2/3, \mu_{\text{D}_2} \approx 1$$

$$E^{\text{vib}} = \left(n + \frac{1}{2} \right) h\nu$$

- The bond strength (k) is the same for H₂, HD et D₂ (covalent bond)
- Vibration energies are different:

$$E_{\text{D}_2}^{\text{vib}} < E_{\text{HD}}^{\text{vib}} < E_{\text{H}_2}^{\text{vib}}$$

- Zero point energy (ZPE=hv/2)



NOTATIONS

For instance for Mg: 3 stable isotopes ^{24}Mg , ^{25}Mg , ^{26}Mg

Isotopic fractionation between two Mg-bearing components A & B:

$$\alpha_{\text{A-B}}^{26/24} = \left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}} \right)_{\text{A}} / \left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}} \right)_{\text{B}}$$

$$\delta^{26}\text{Mg}_{\text{A}} = \left(\frac{\left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}} \right)_{\text{A}}}{\left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}} \right)_{\text{Standard}}} - 1 \right) \times 1000 \quad \Delta_{\text{A-B}}^{26/24} = \delta^{26}\text{Mg}_{\text{A}} - \delta^{26}\text{Mg}_{\text{B}} \approx 1000 \times \ln \left(\alpha_{\text{A-B}}^{26/24} \right)$$

TWO TYPES OF ISOTOPIC FRACTIONATIONS

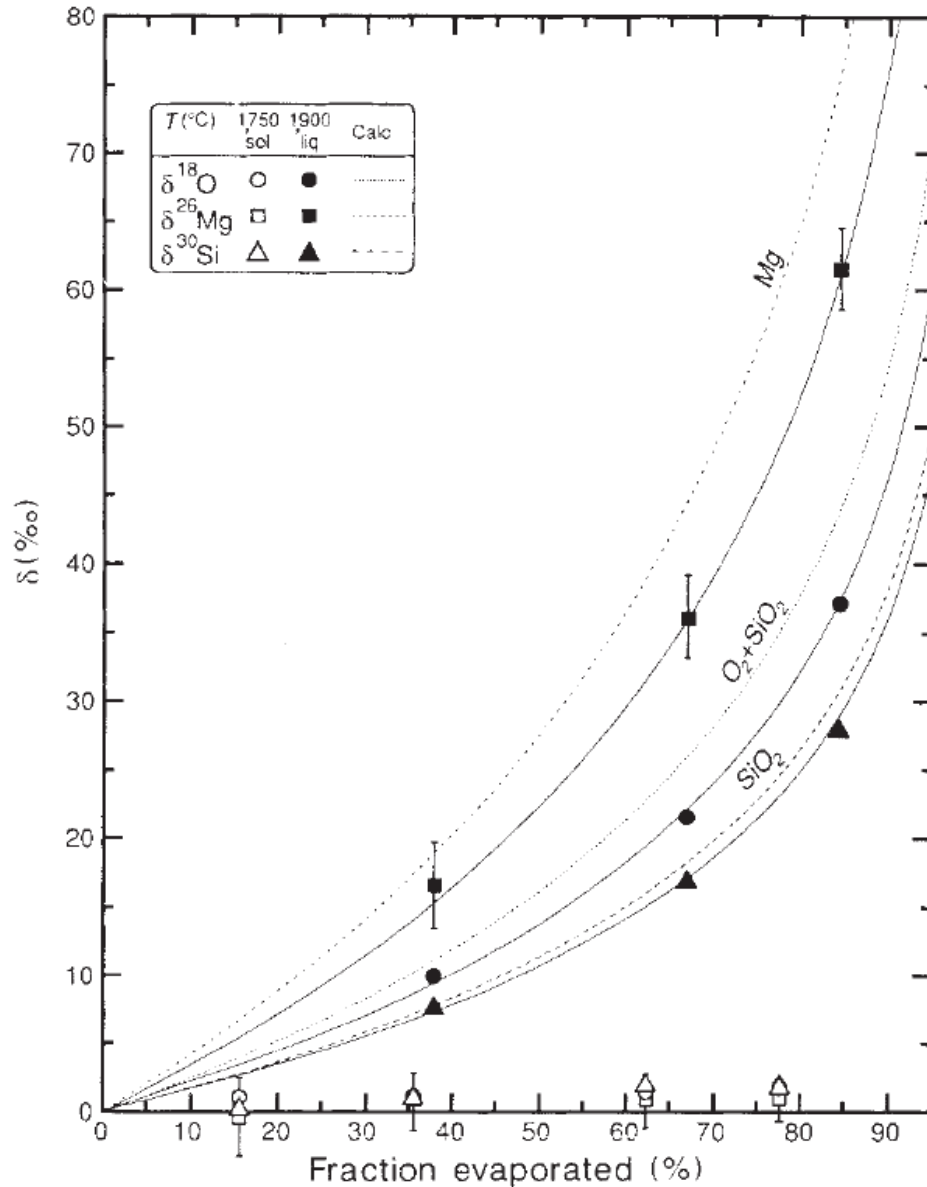
- Equilibrium isotopic fractionations: the isotopes are distributed to minimize the total energy of the system.
- Kinetic isotopic fractionations: transport properties (diffusion) of isotopes of a given element are different (for irreversible reactions, e. g. evaporation, condensation, ...)

These two isotopic fractionations are mass dependent: $\alpha_{\text{A-B}}^{25/24} = \left(\alpha_{\text{A-B}}^{26/24} \right)^{\beta}$

Equilibrium $\beta = \left(\frac{1}{24} - \frac{1}{25} \right) / \left(\frac{1}{24} - \frac{1}{26} \right) = 0.521$

Kinetic $\beta = \ln \left(\frac{24}{25} \right) / \ln \left(\frac{24}{26} \right) = 0.511$

Experimental determinations of the O, Si and Mg isotopic fractionations during evaporation of molten Mg_2SiO_4



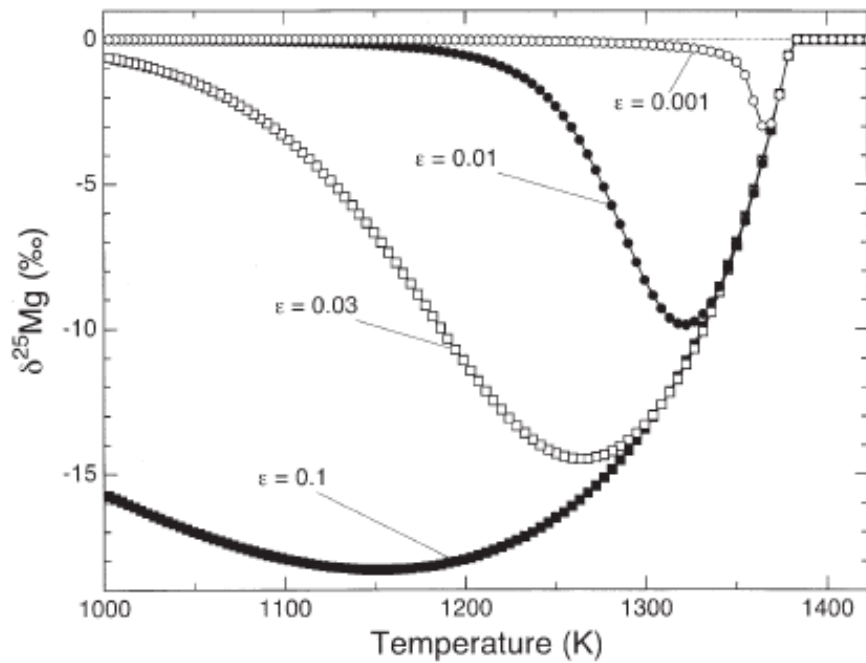
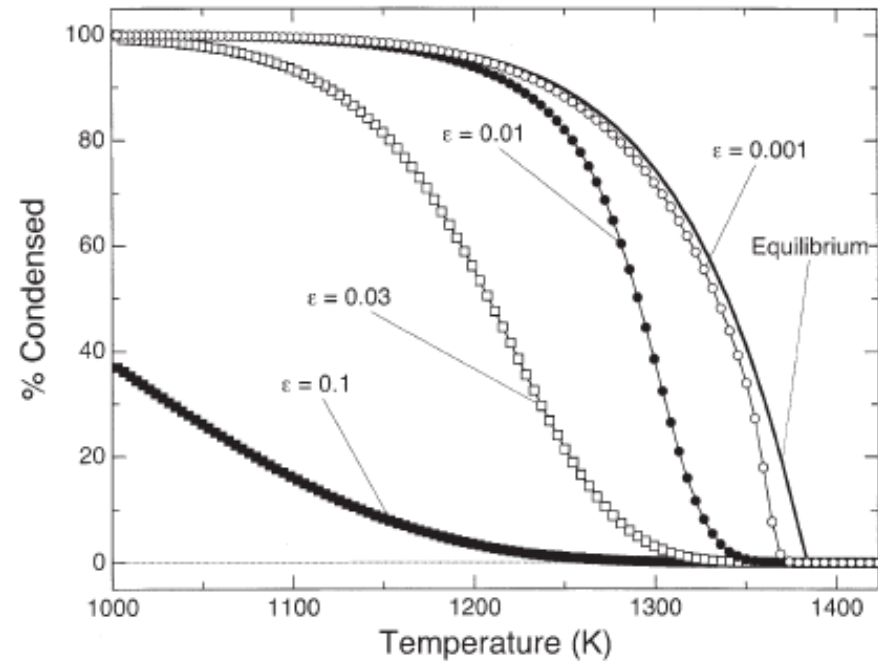
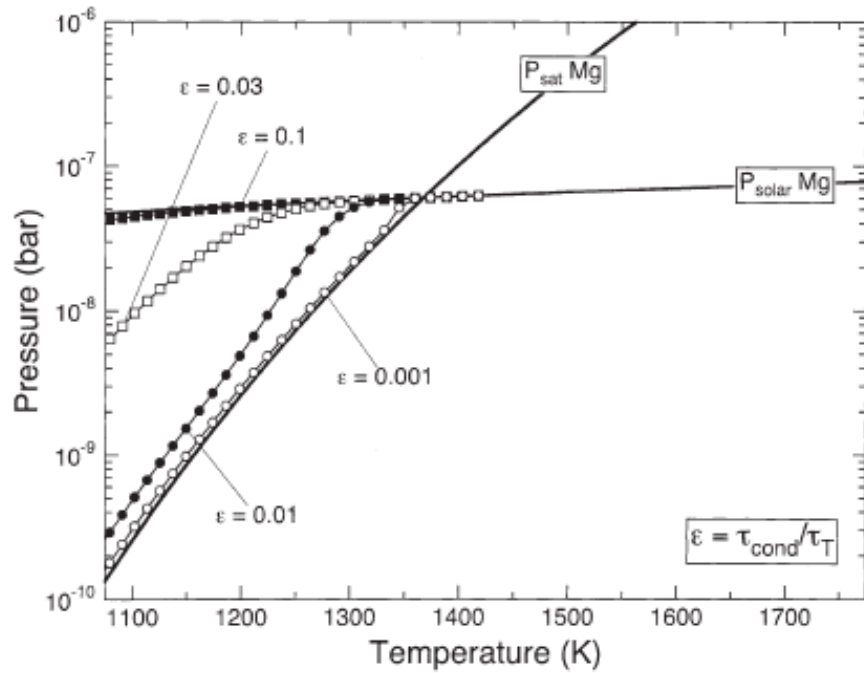
Davis et al., 1990

Enrichments in heavy isotopes of the residues of evaporation according to a Rayleigh mass balance:

$$R = R_{\text{initial}} \times f^{(\alpha-1)}$$

With α depending on the species evaporated:
e.g. for Mg isotopes if Mg evaporates as MgO :

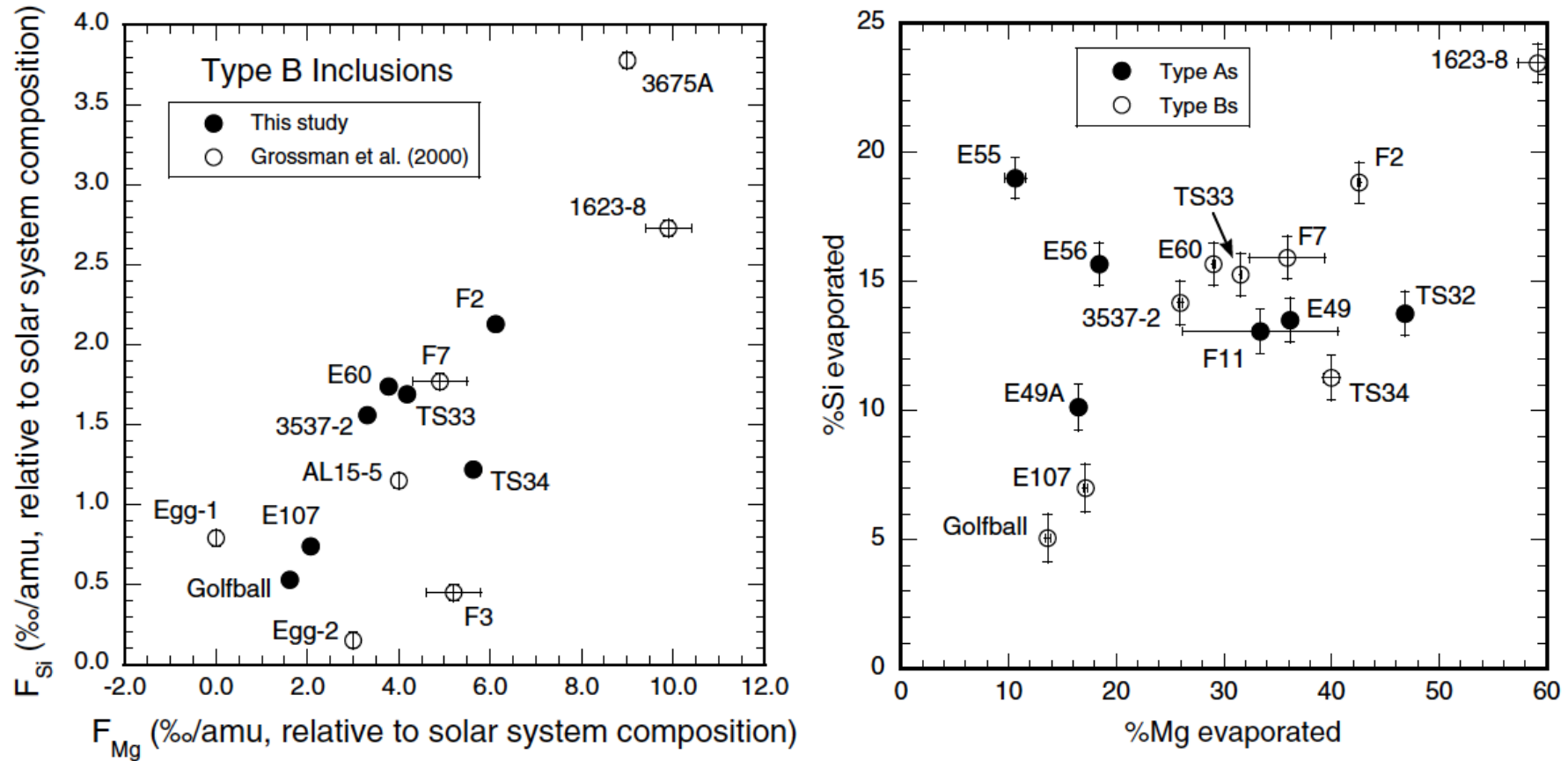
$$\alpha_{\text{gas-molten olivine}}^{26/24} \approx \sqrt{(24+16)/(25+16)} = 0.988$$



Large isotopic fractionations (here Mg) can also be produced in case of non-equilibrium condensation ($\epsilon \ll 1 =$ slow cooling)

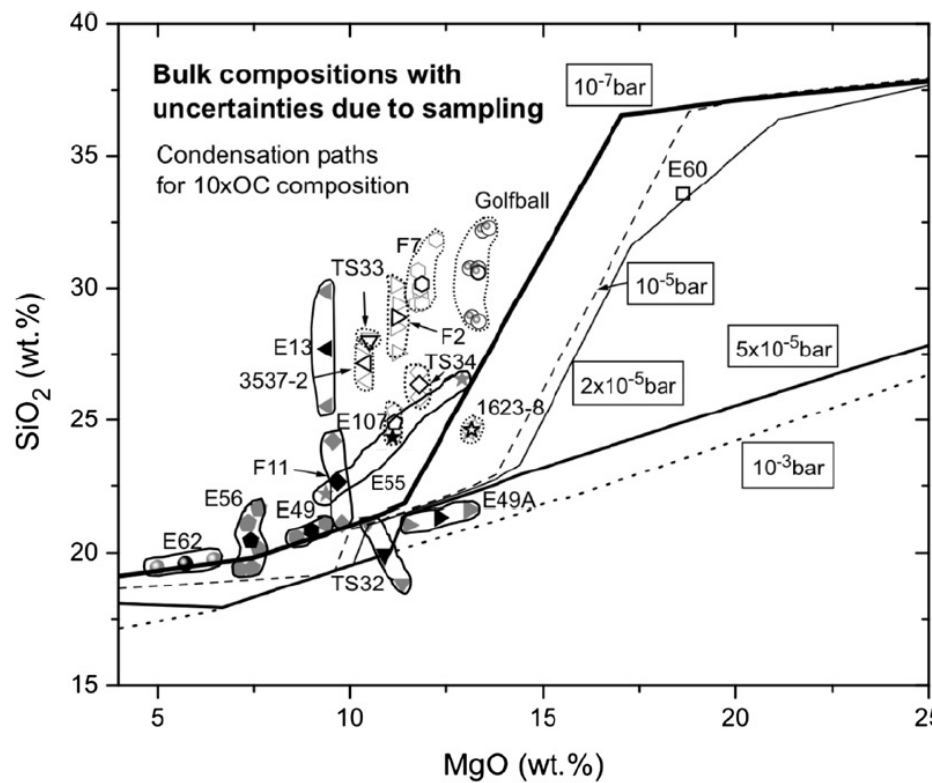
Richter, 2004

Enrichments of the O, Si and Mg isotopic compositions of CAIs can be used to determine the fraction (1-f) of O, Mg and Si evaporated

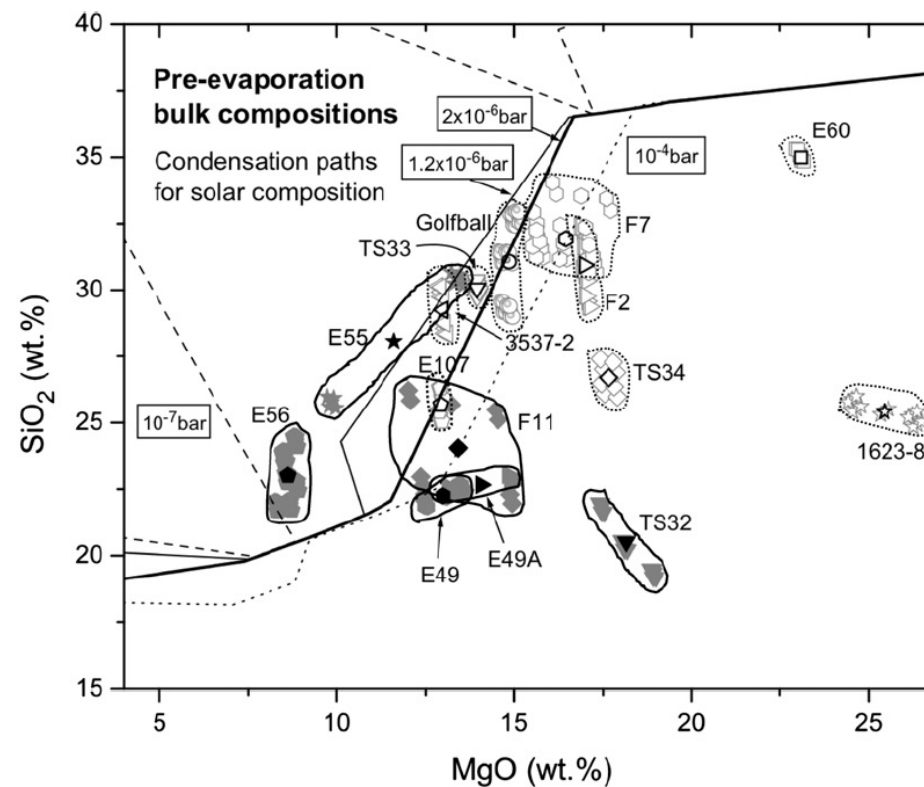


Grossman et al., 2008 (see also Mendybaev et al., 2013 for FUN CAIs)

*Before correction for evaporation loss
of Mg and Si*

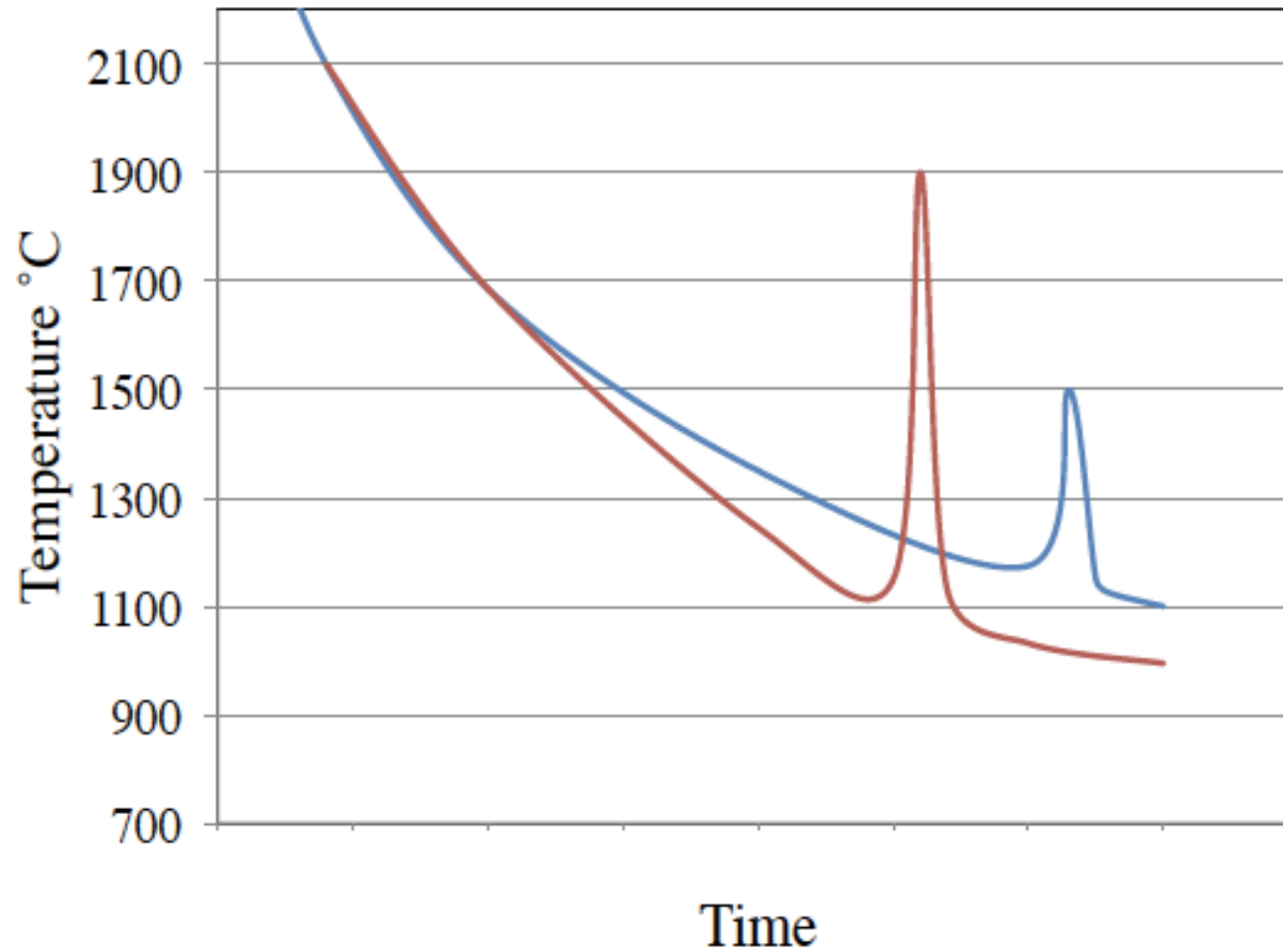


*After correction for evaporation loss
of Mg and Si*



Grossman et al., 2008

CAIs had a complex history from the condensation of their precursors to later melting and evaporation events



Courtesy Frank Richter

October 26th to 31st

Labex **UnivEarthS**

FALL SCHOOL 2014

Villa Finaly, Florence (Italy)



- How (to first order) did the Solar system and the planets form?
What are primitive meteorites?
- The links between the bulk chemical and isotopic compositions of the Earth and that of the Sun (and of the galaxy)
Earth = CMAS composition (Ca+Mg+Al+Si, +O and also Fe)
- What is the origin of solids in the Solar system?
Fractionation (chemical and isotopic) processes during condensation/evaporation in the Solar protoplanetary disk: from the gas to the dust
Variations of Al/Si and Mg/Si ratios and isotopic compositions of O, Mg and Si
- Timing of condensation and accretion in the Solar protoplanetary disk: from the dust to the building blocks of the Earth
Radioactive decay of short-lived ^{26}Al into ^{26}Mg



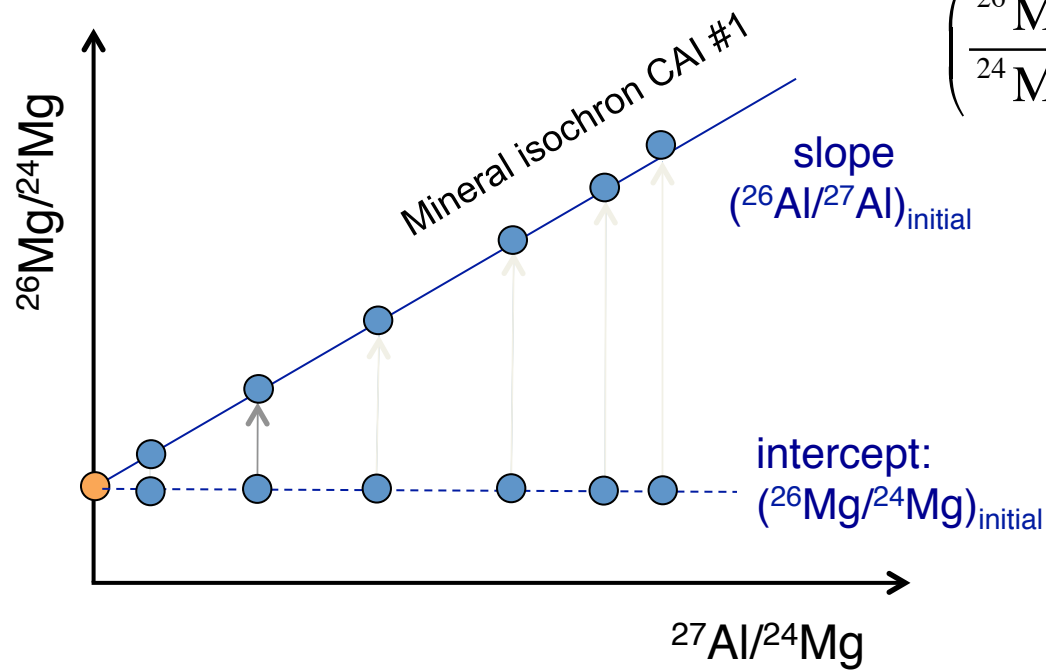
The questions

- When and how fast did the components of chondrites (CAIs and chondrules) and their precursors form?
- When did the rock itself (the chondrite) form?
- Is it a sediment of “first generation” made from dust derived directly from the nebula gas or of “second generation” made from fragments of pre-existing planets?
- How do these ages for the formation and evolution of dust compare with ages observed for young stars analogues to the Sun?

How to do it

- Need for a chronometer which could work for fast processes (short half-life), date condensation-melting-metamorphism (fractionation of parent/daughter ratio): short-lived radioactive nuclides (^{10}Be , ^{26}Al , ^{41}Ca , ^{53}Mn , ^{60}Fe , ^{182}Hf) of “last minute” origin.

The presence of ^{26}Al at the time of crystallisation of an object is demonstrated by the fact that the ^{26}Mg excesses are correlated with the $^{27}\text{Al}/^{24}\text{Mg}$ ratios
(Lee, Papanastassiou & Wasserburg, 1976)



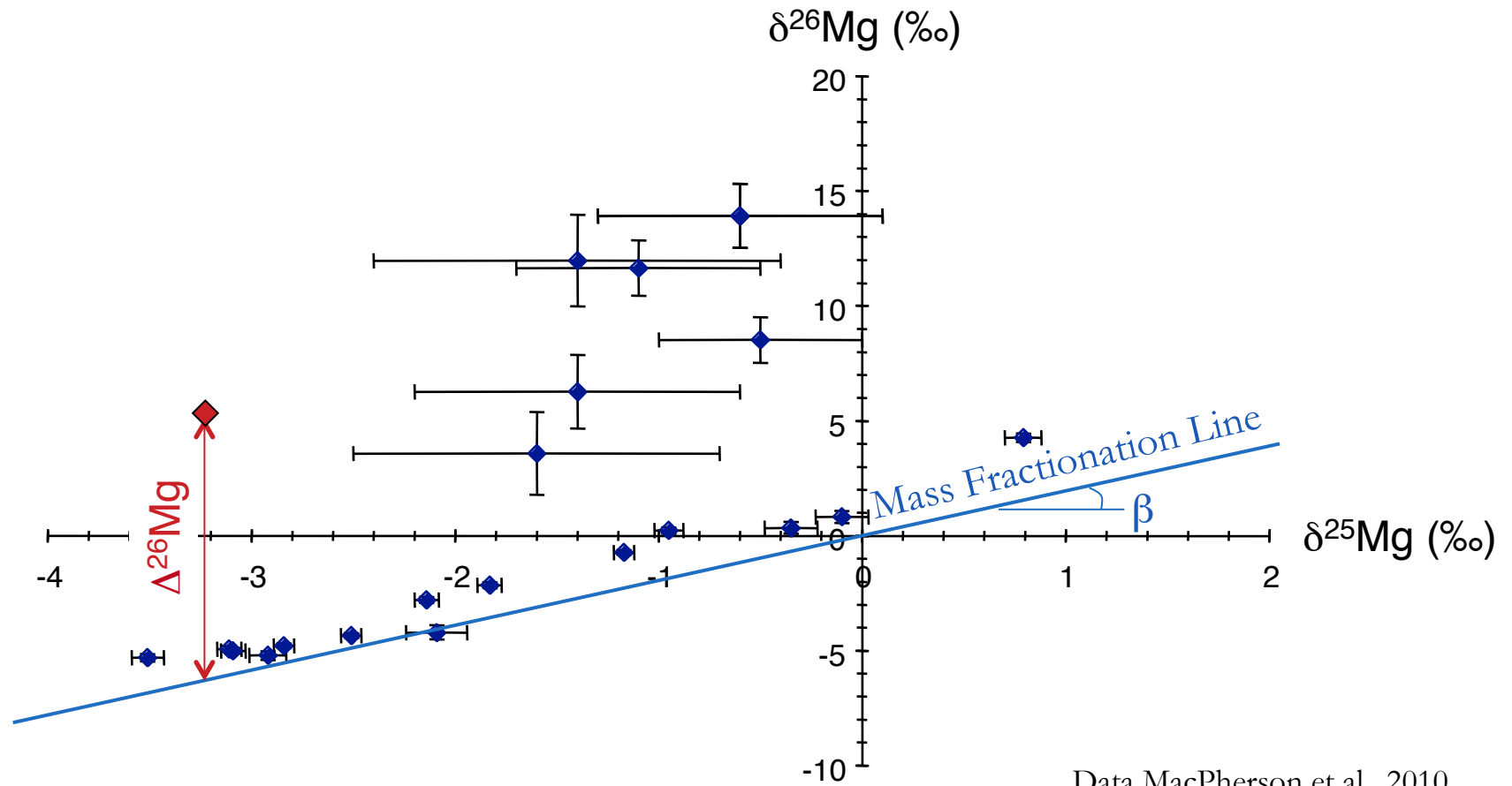
$$\left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}}\right) = \left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}}\right)_{\text{initial}} + \left(\frac{^{26}\text{Al}}{^{27}\text{Al}}\right)_{\text{initial}} \times \left(\frac{^{27}\text{Al}}{^{24}\text{Mg}}\right)$$

Two objects with different initial $^{26}\text{Al}/^{27}\text{Al}$ (at the time of their crystallization) will give different mineral isochrons.

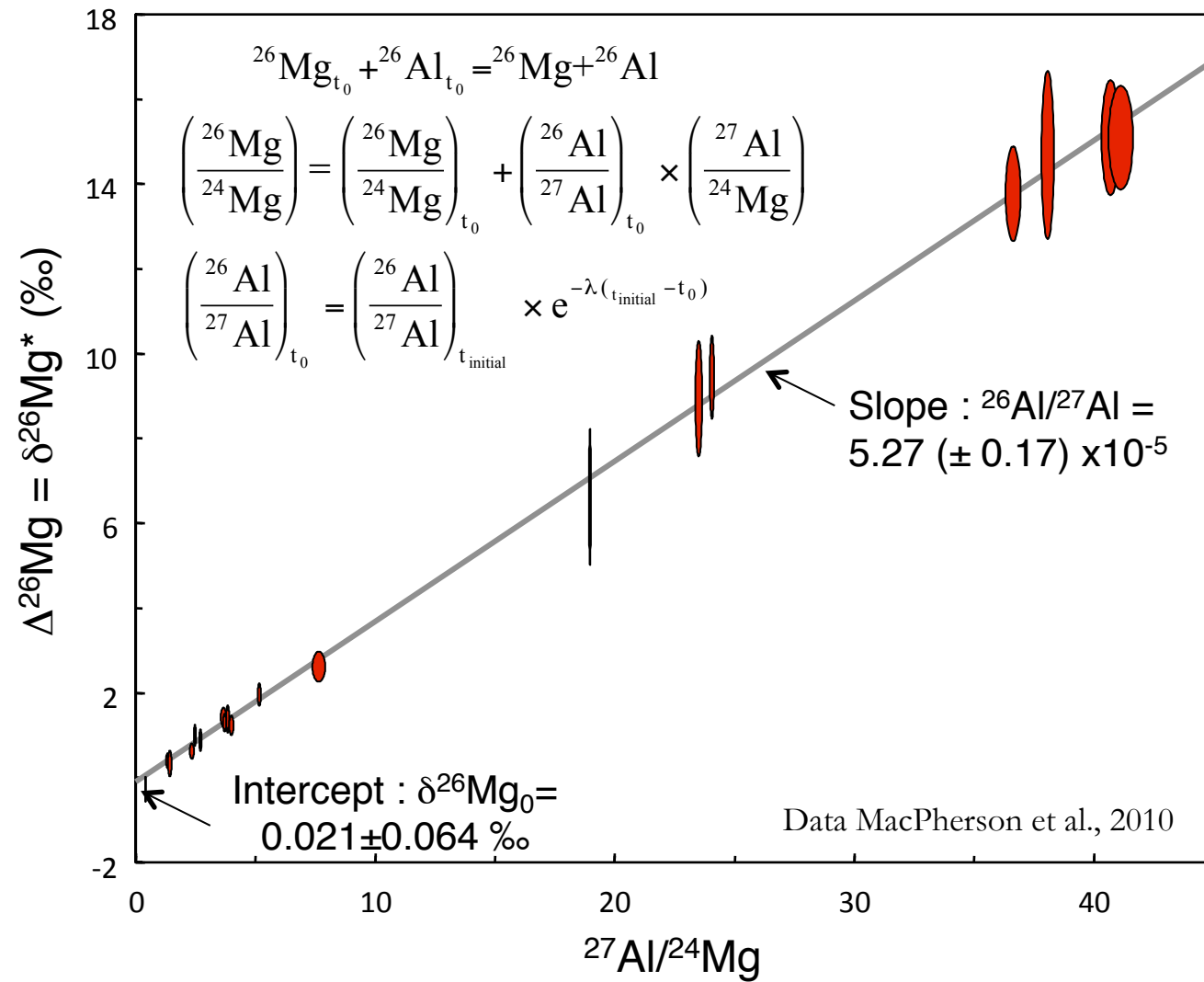
$$\left(\frac{^{26}\text{Al}}{^{27}\text{Al}}\right)_{\text{initial}} = \left(\frac{^{26}\text{Al}}{^{27}\text{Al}}\right)_0 \times e^{-\lambda(t_0 - t_{\text{initial}})}$$

The difference in their $^{26}\text{Al}/^{27}\text{Al}$ ratios can be converted into time difference if and only if it can be shown that they derive from a common reservoir once homogenized (to some level) in $^{26}\text{Al}/^{27}\text{Al}$

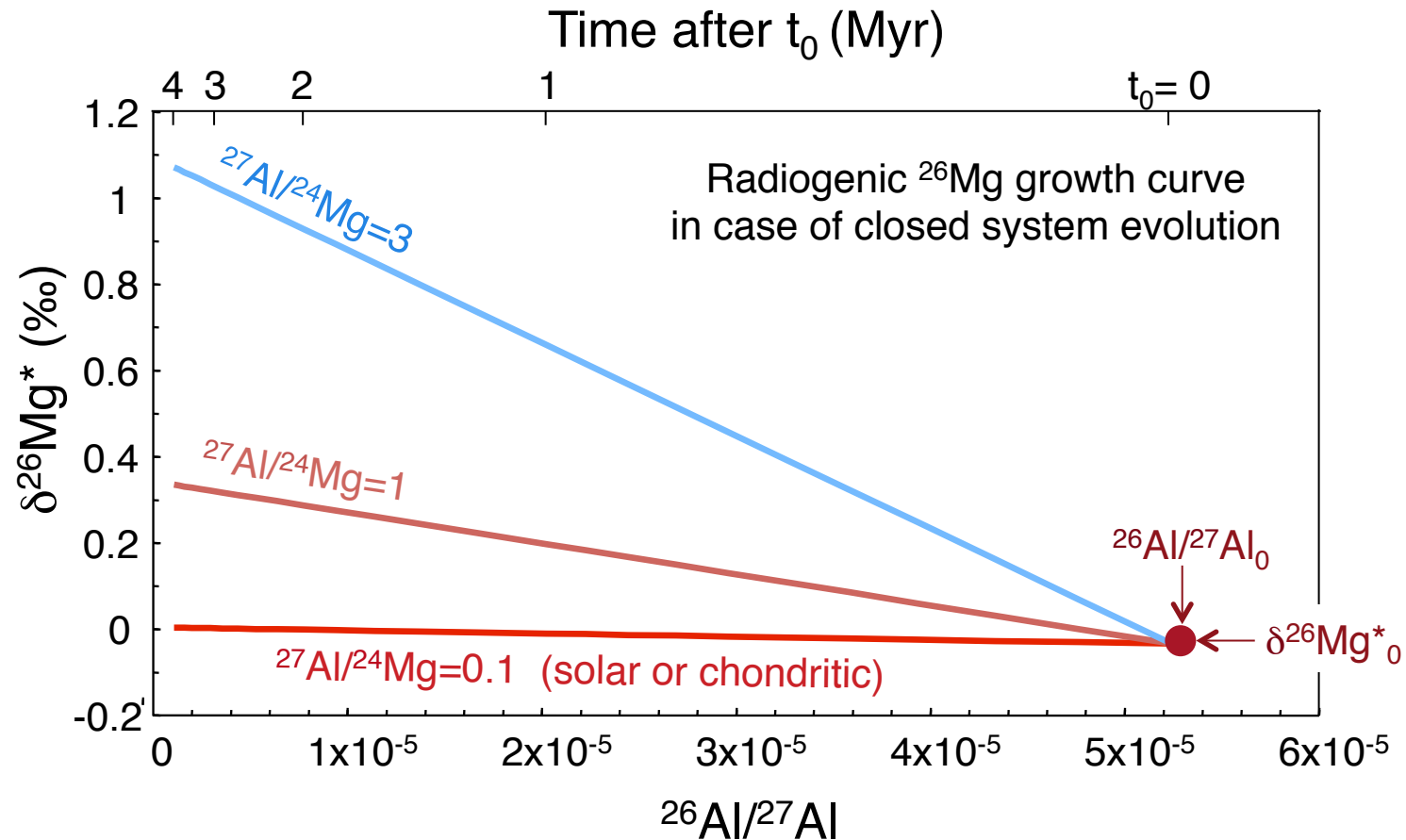
^{26}Mg excesses ($\Delta^{26}\text{Mg}$) in Ca-, Al-rich refractory inclusions



The ^{26}Mg excesses are due to the in situ decay of short-lived ^{26}Al

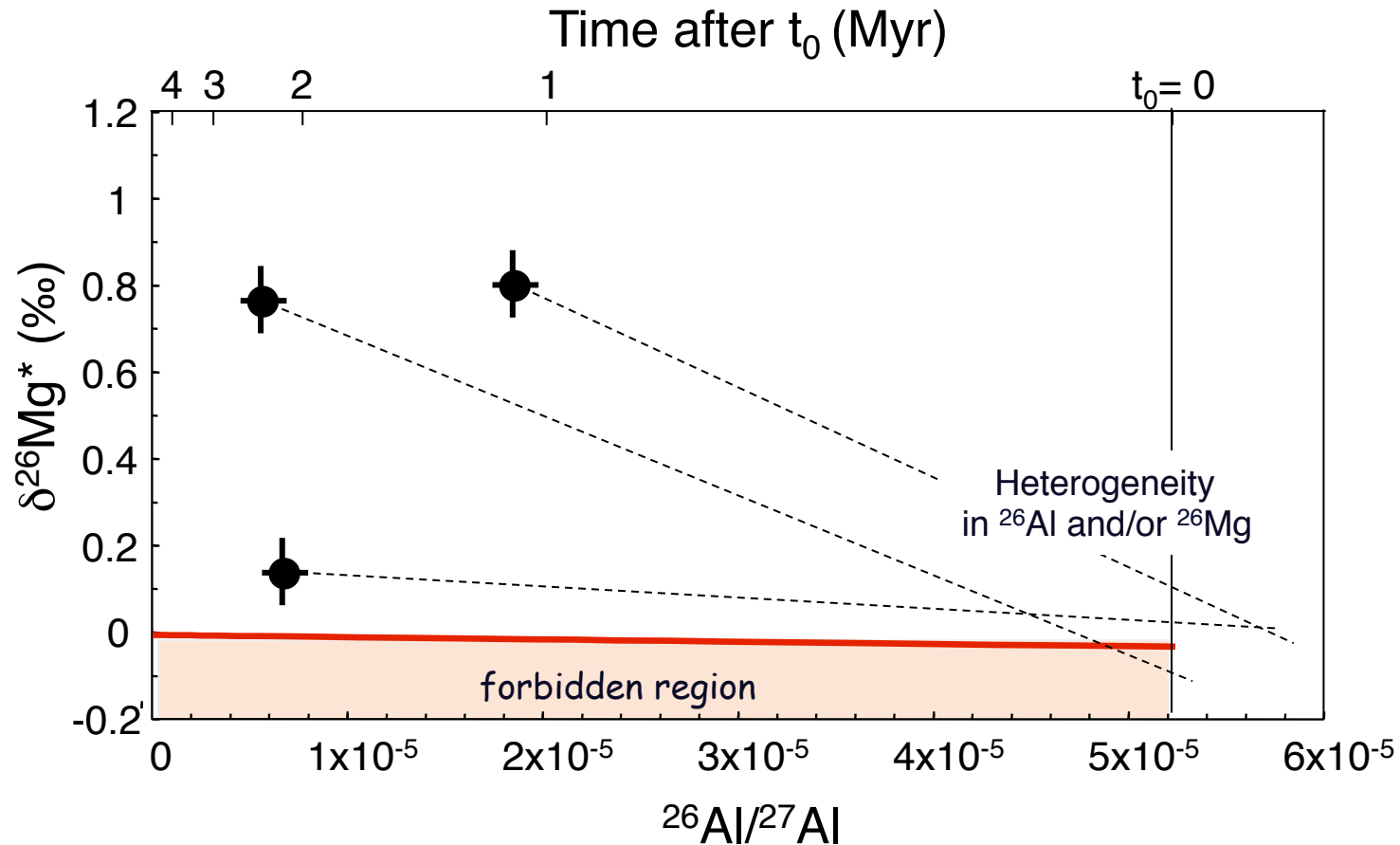


Not only the slope but also the initial of the ^{26}Al isochron carry an information on age

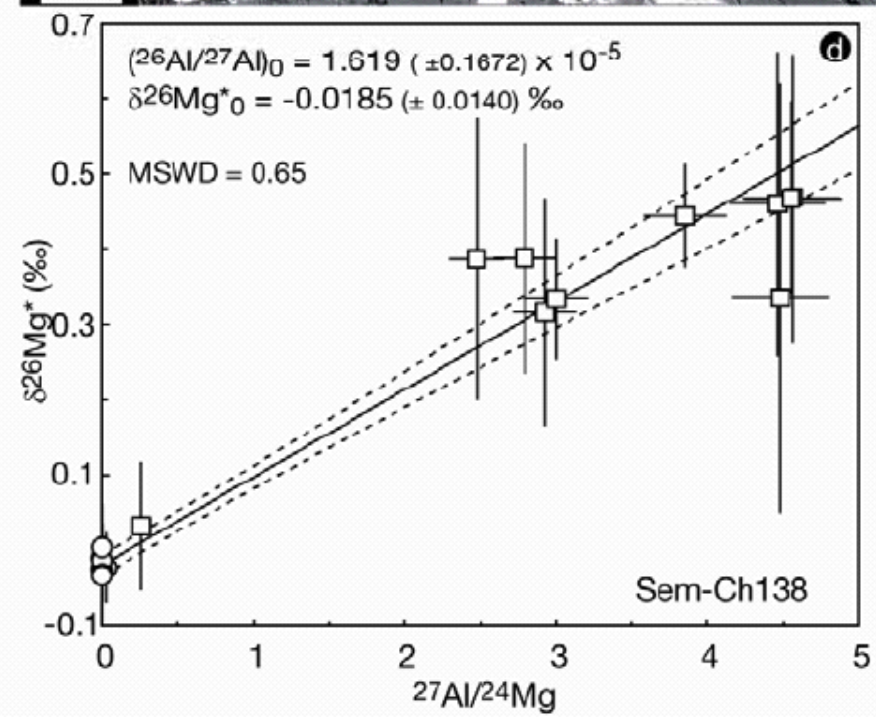
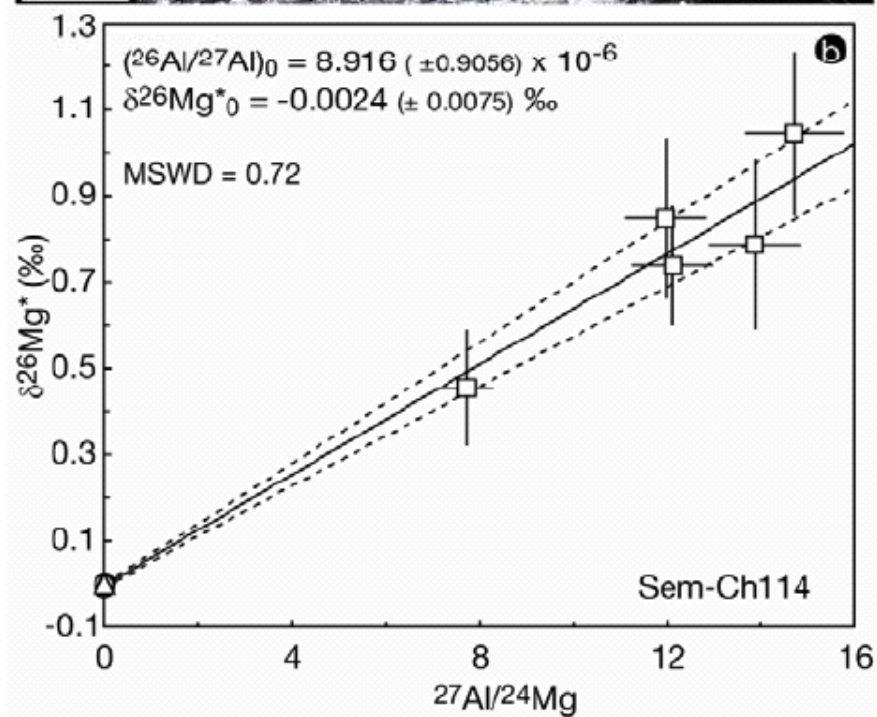
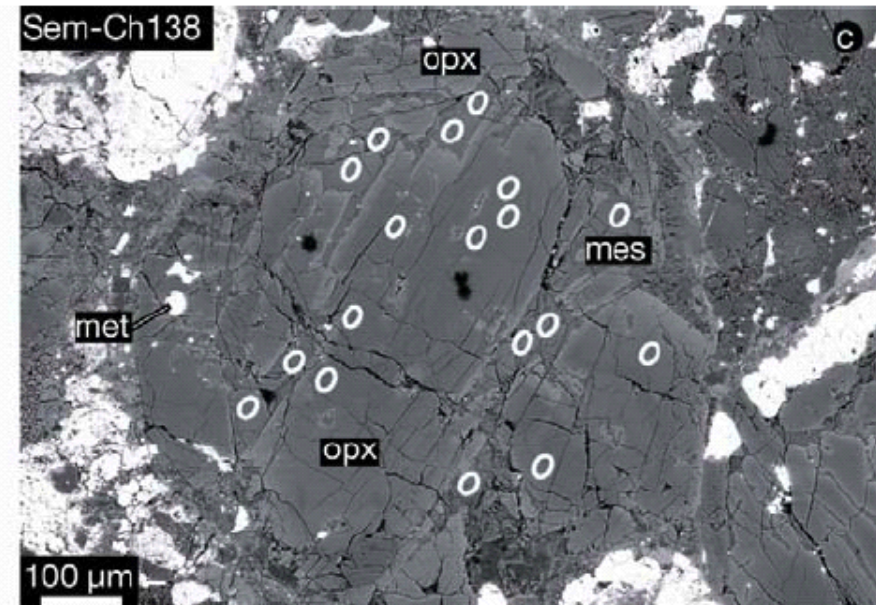
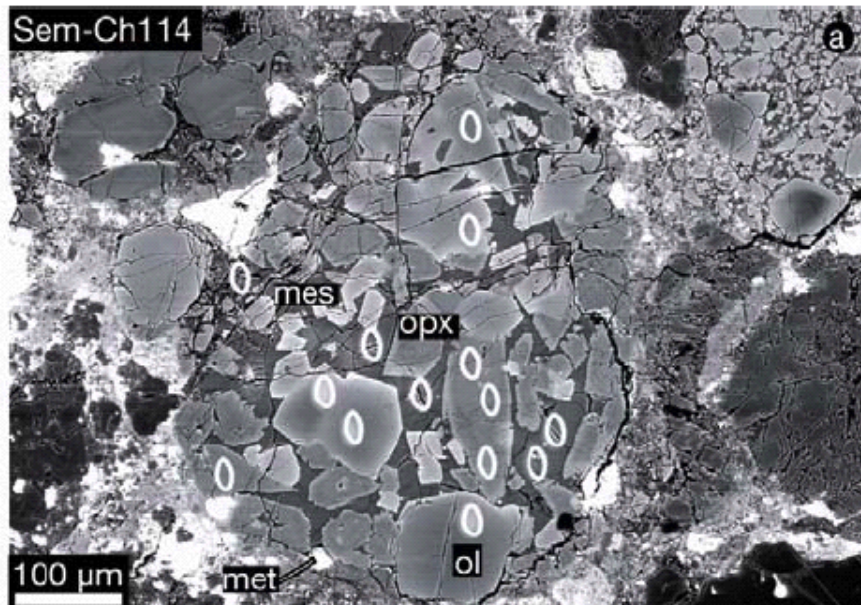


The increase of $^{26}\text{Mg}/^{24}\text{Mg}$ is of 38 ppm (0.038 ‰ in $\delta^{26}\text{Mg}$ unit) for a solar $^{27}\text{Al}/^{24}\text{Mg}$ ratio

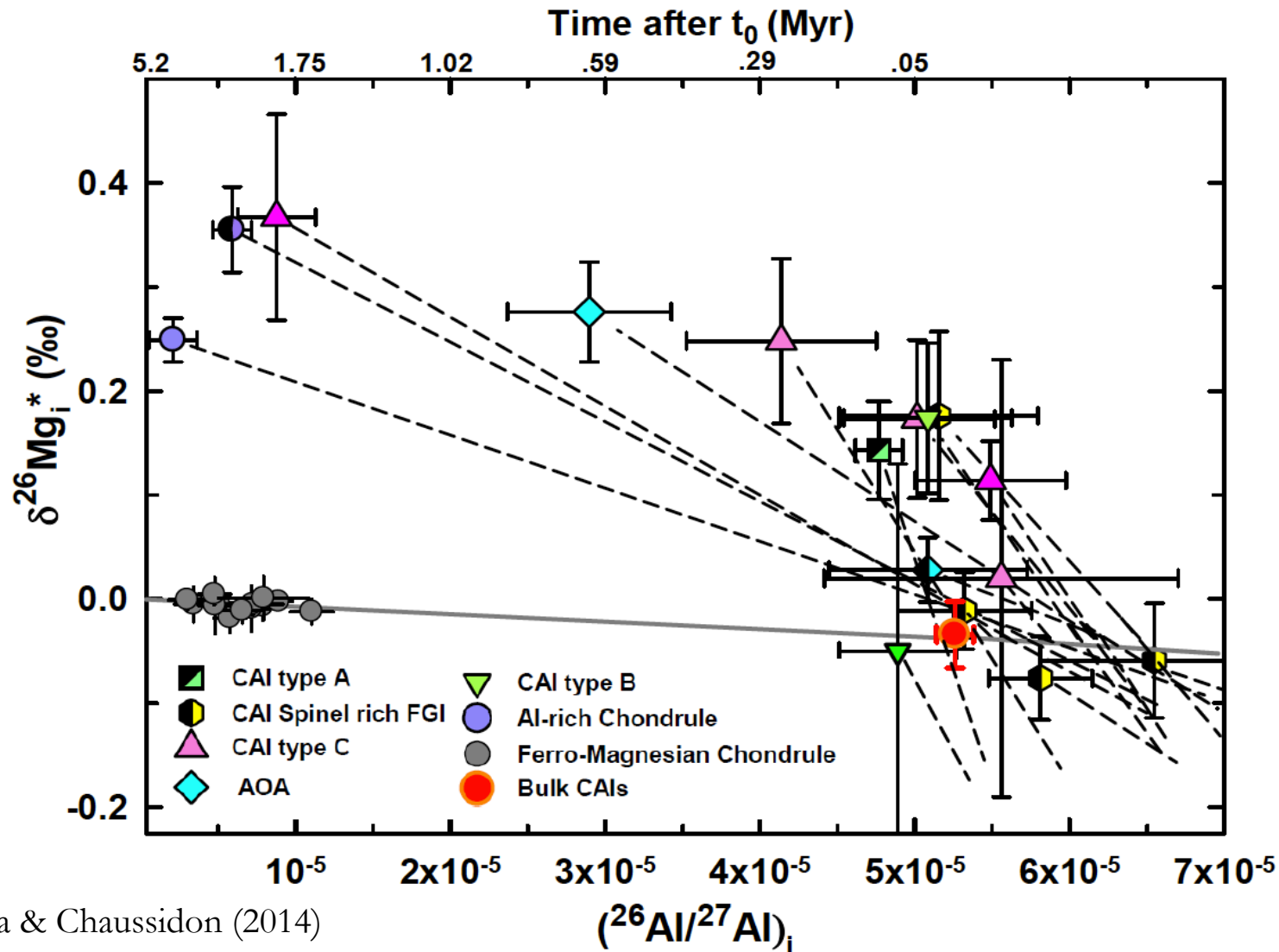
The initial $\delta^{26}\text{Mg}^*$ and $^{26}\text{Al}/^{27}\text{Al}$ from the isochron can be used to constrain the homogeneity of the distribution of Al and Mg isotopes in the reservoir parent to CAIs and chondrules (Villeneuve et al., 2009; Larsen et al., 2011; MacPherson et al., 2012)



$$\delta^{26}\text{Mg}^*_{\text{initial}} = \delta^{26}\text{Mg}^*_0 - \frac{^{27}\text{Al}/^{24}\text{Mg}}{\left(^{26}\text{Mg}/^{24}\text{Mg}\right)_{\text{ref}}} \times \left[\left(^{26}\text{Al}/^{27}\text{Al}\right)_0 - \left(^{26}\text{Al}/^{27}\text{Al}\right)_{\text{initial}} \right] \times 1000$$

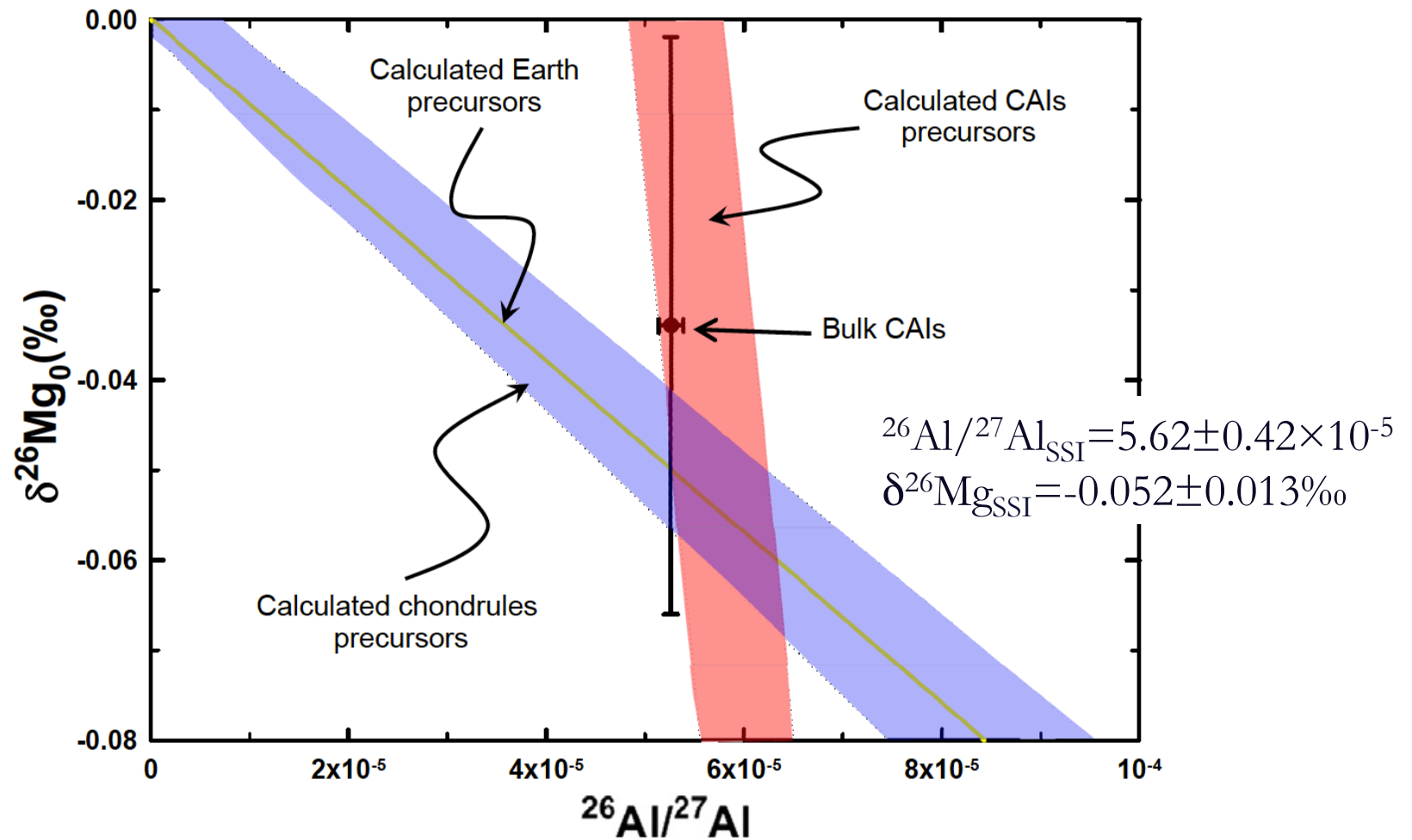


CAIs and chondrules from different chondrites, despite having different ^{26}Al ages (up to $\approx 3\text{-}4$ Myrs range) and $^{27}\text{Al}/^{24}\text{Mg}$ ratios, can all derive from precursors condensed early from the same gas (for Mg and Al isotopes) (discordant view in Larsen et al., 2011)

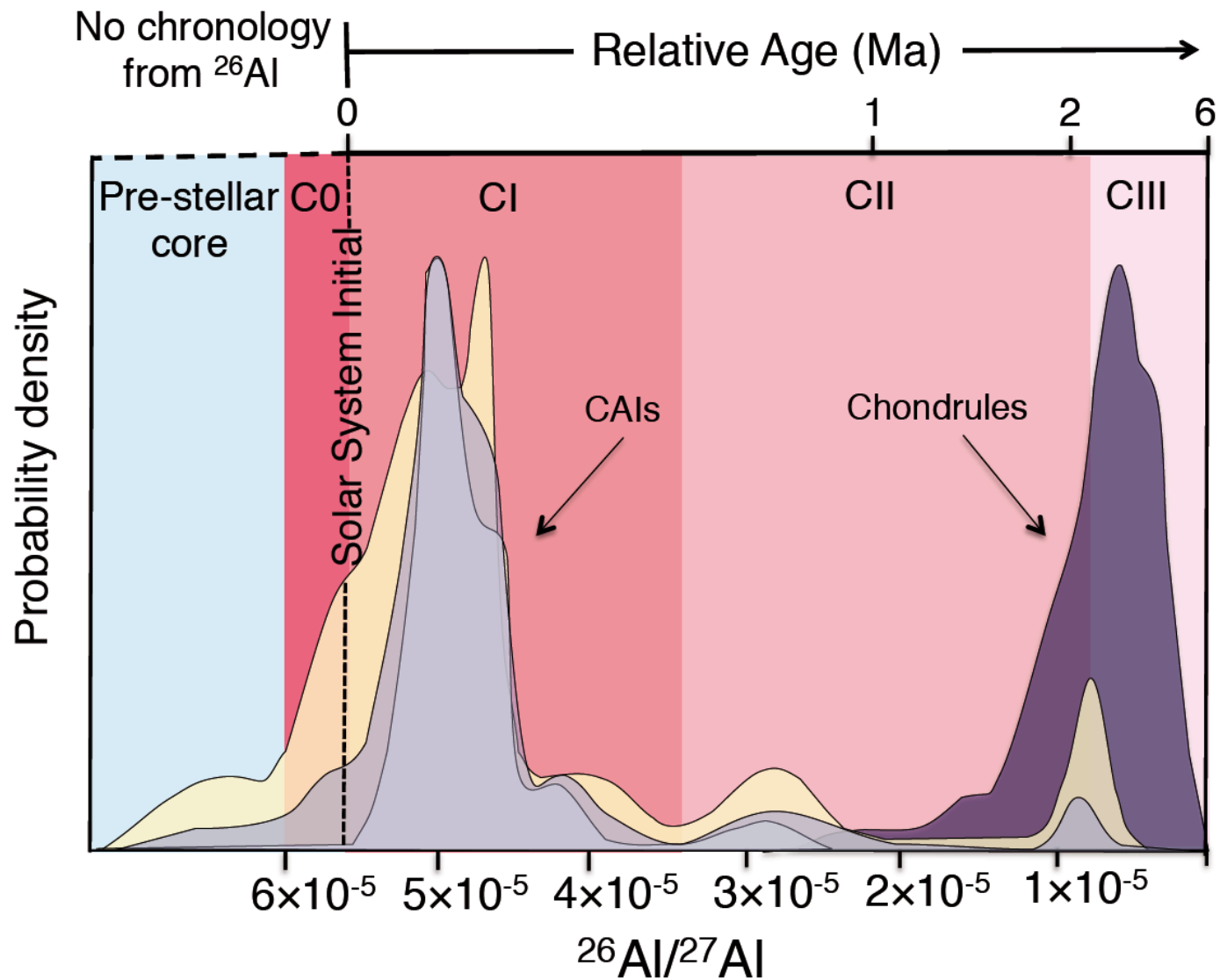


Mishra & Chaussidon (2014)

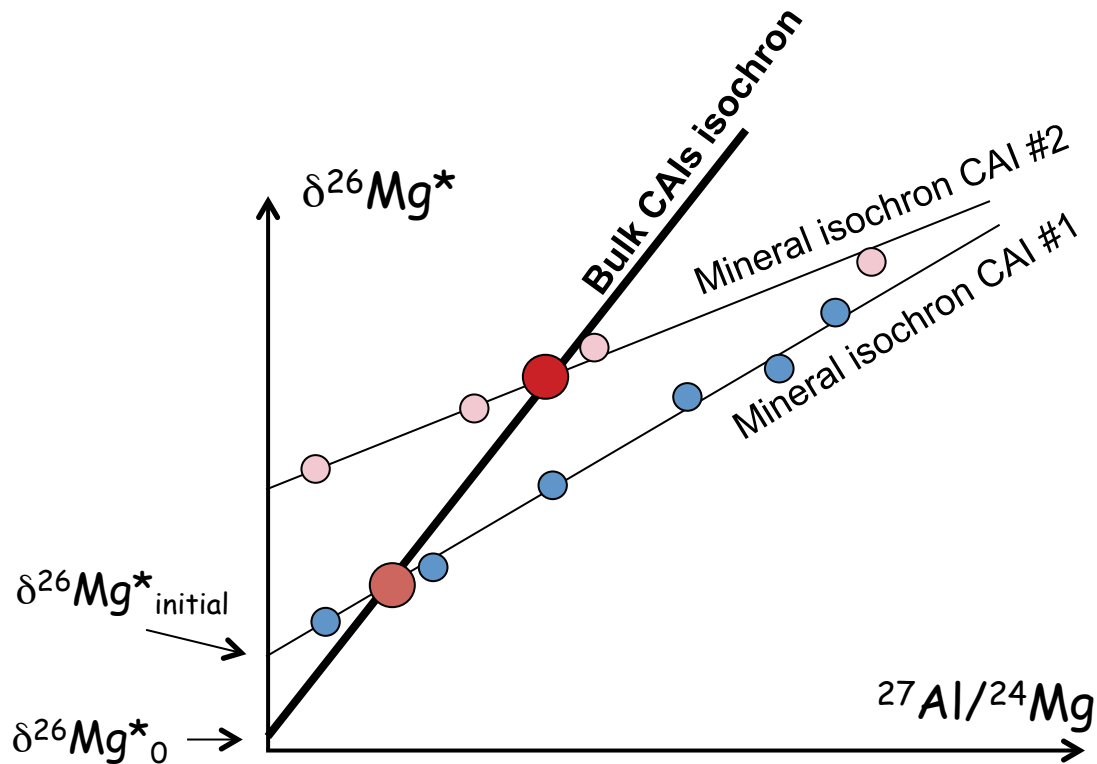
Because ferro-magnesian chondrules and refractory objects have very different $^{27}\text{Al}/^{24}\text{Mg}$ ratios, the composition calculated for their precursors has a well defined intersection (Solar System Initial)



The chronology of the early Solar System reconstructed from ^{26}Al studies of CAIs and chondrules is in line with the time evolution of young stars

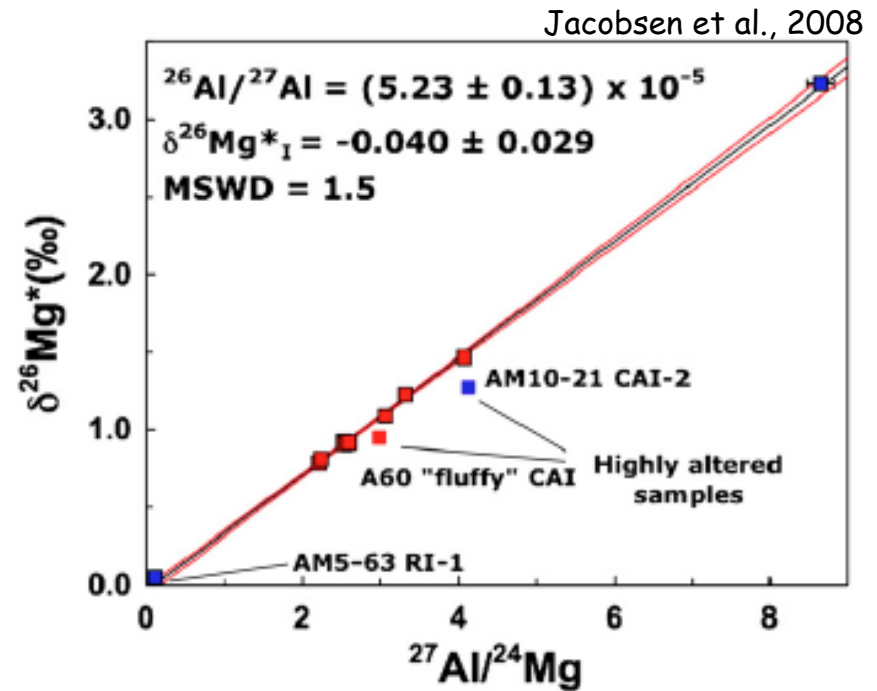
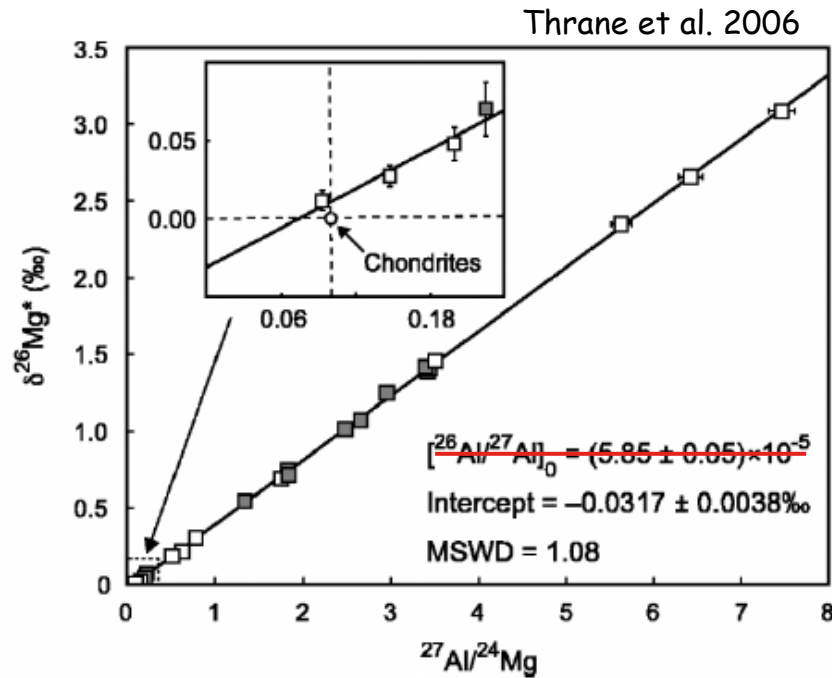


How to go a step further: when did the first dust form?
When did the precursors of CAIs and chondrules form?



- Al/Mg fractionation for mineral isochrons:
crystal/liquid partitioning during magmatic history of CAIs or chondrules
- Al/Mg fractionation for bulk isochrons:
precursors composition, condensation & evaporation of CAI or chondrule melts

High precision MC-ICPMS analyses of bulk CAIs from CV chondrites show that they define a very tight bulk ^{26}Al isochron
 (Thrane et al. 2006 ; Jacobsen et al. 2008; Larsen et al. 2011)

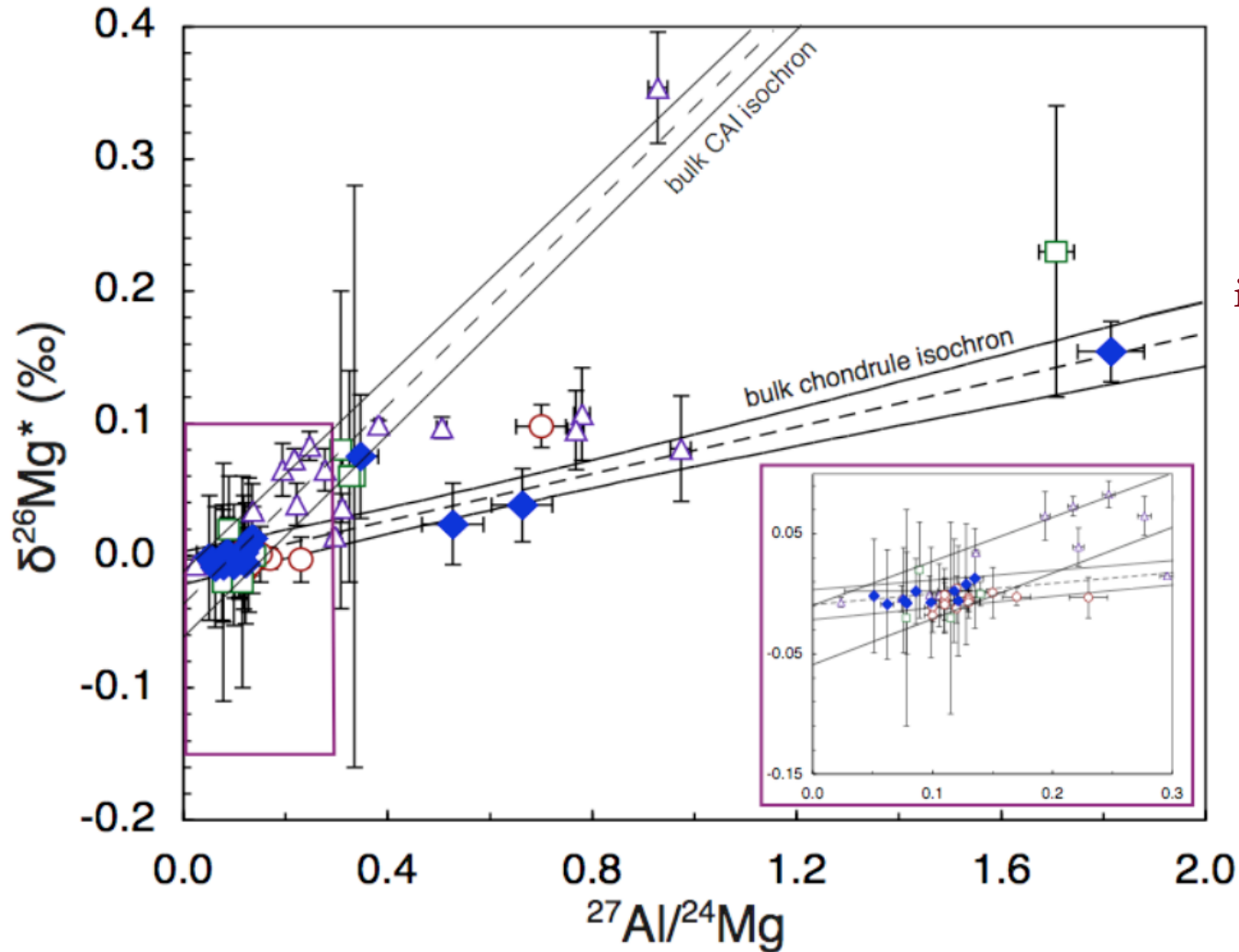


Very short interval for condensation
 $\pm 0.05 \times 10^{-5} \longleftrightarrow \pm 10,000$ years

Questions: only one event which lasted 10 000 years (or even 3000 years, Larsen et al., 2011), or many events within 10 000 years, or formation of CAIs over a much longer period but bias from sample selection ?

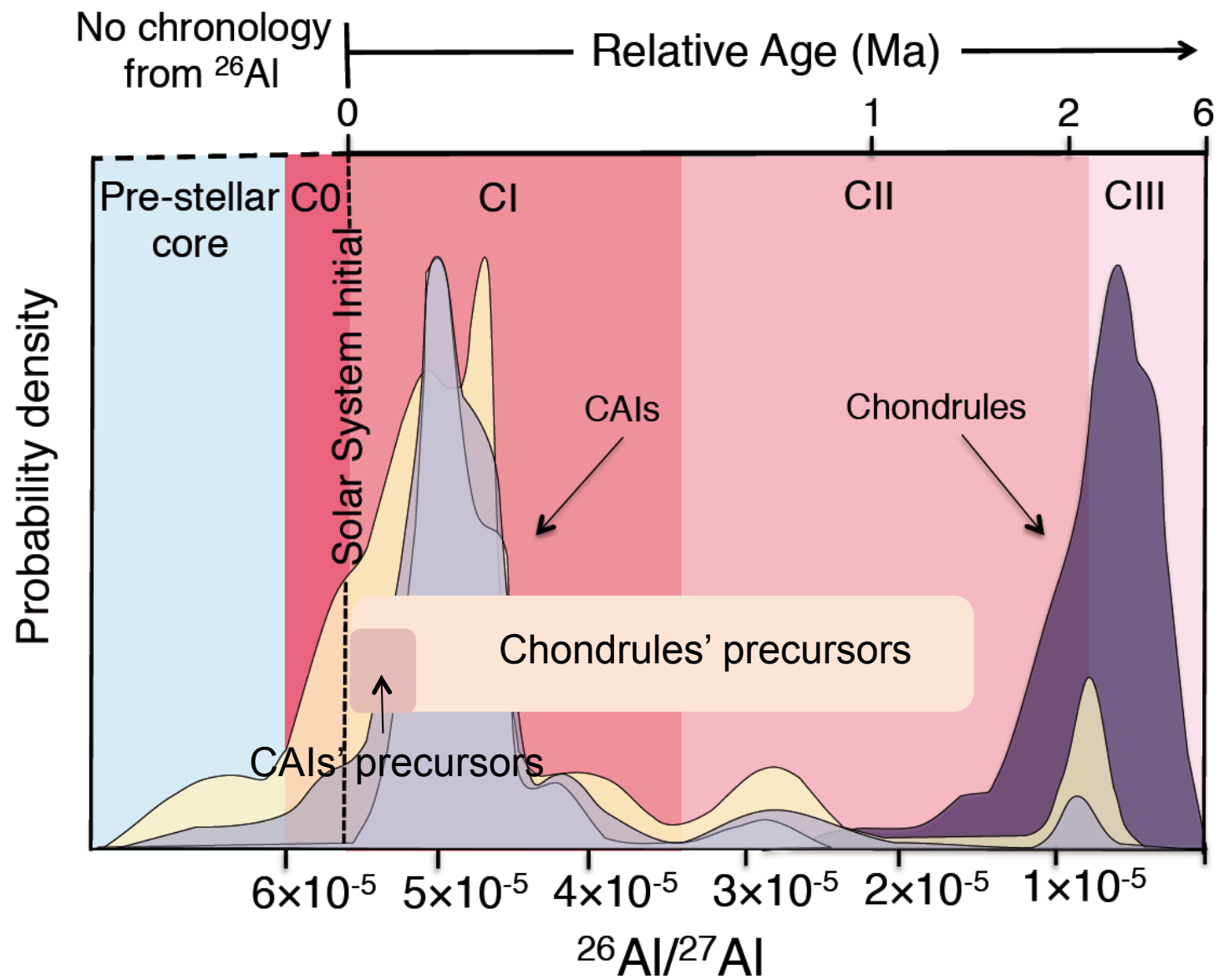
The bulk CAIs' isochron marks the end of the high-temperature evolution of their precursors in the gas

No contradiction: chondrule precursors formed from time zero during 1.5 Myr.
No chondrule precursor formed later. Some chondrules were formed/remelted later,
up to 3-4 Myr later.



The bulk chondrules' isochron marks the end of the formation and evolution of their precursors in the gas

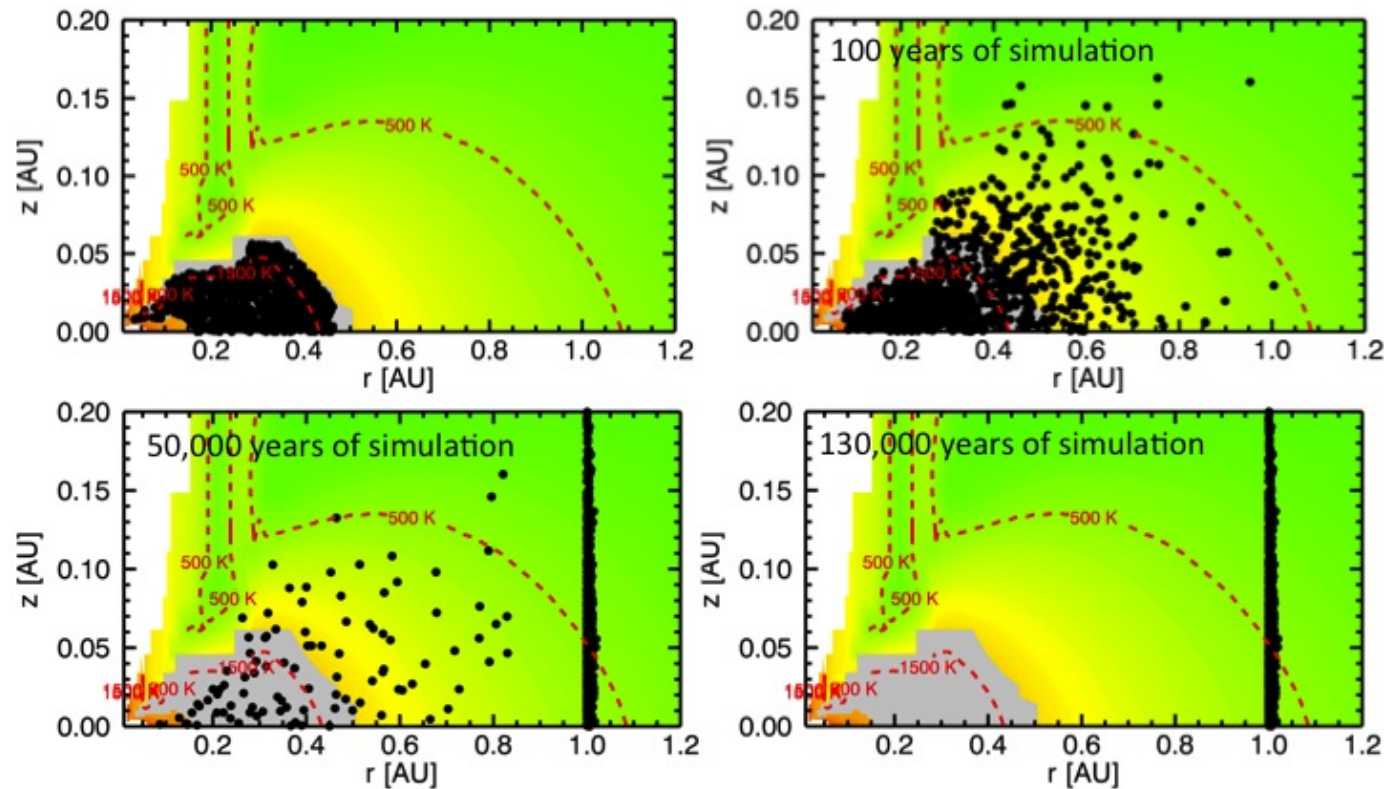
Data from Galy et al., 2000; Bizzarro et al., 2004;
Villeneuve et al., 2009; Luu et al. in prep)





Many questions (or constraints) on transport processes in the accretion disk and residence time of grains:

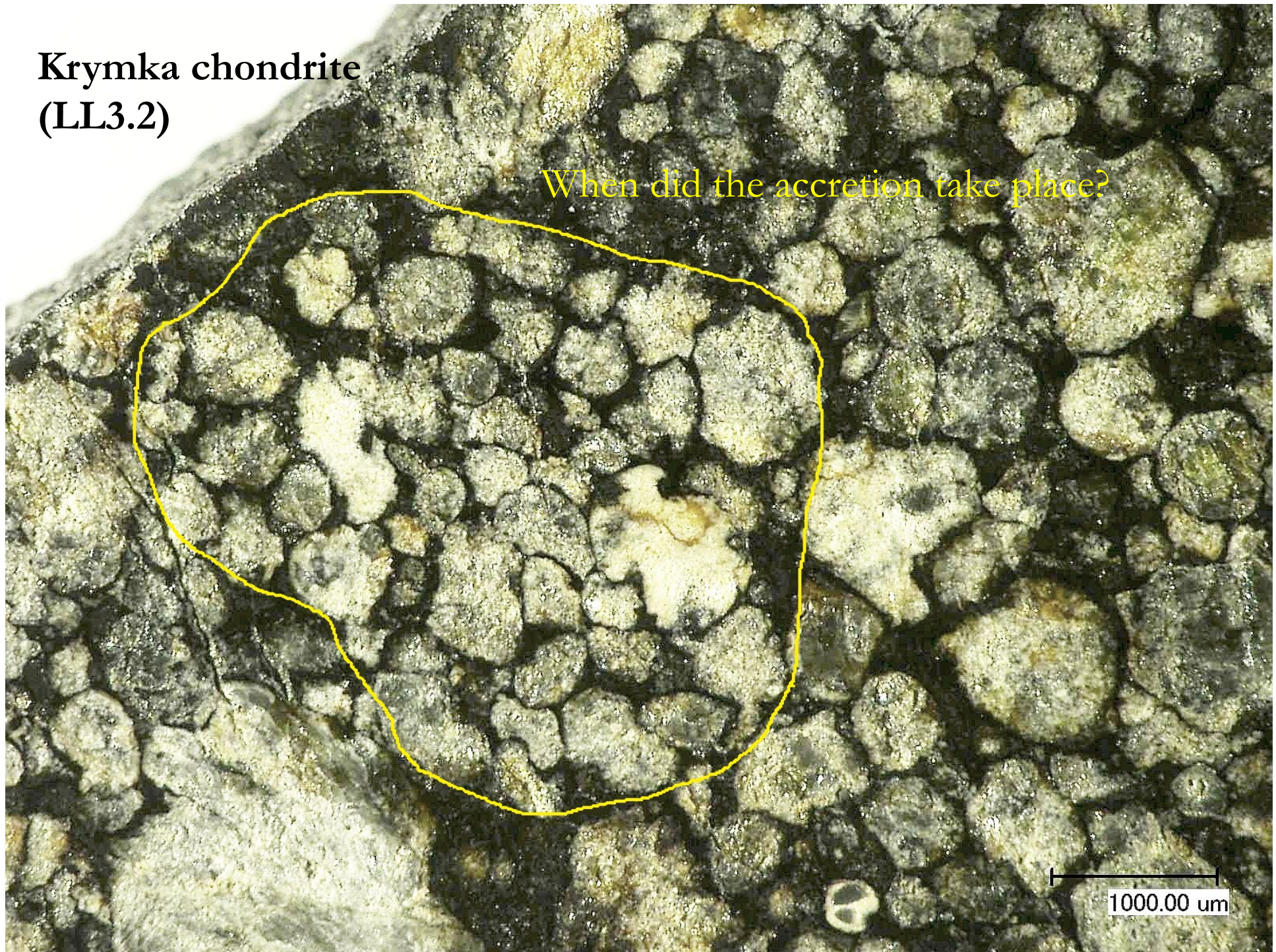
- how to preserve only the earliest formed CAIs in chondrites (viscous dissipation in the disk, Ciesla 2010)
- how to obtain residence times of up to 1.5 Myr (or even 3 Myr) for the grains (early formation of larger aggregates, Ciesla 2007; dead-zone)
- ...



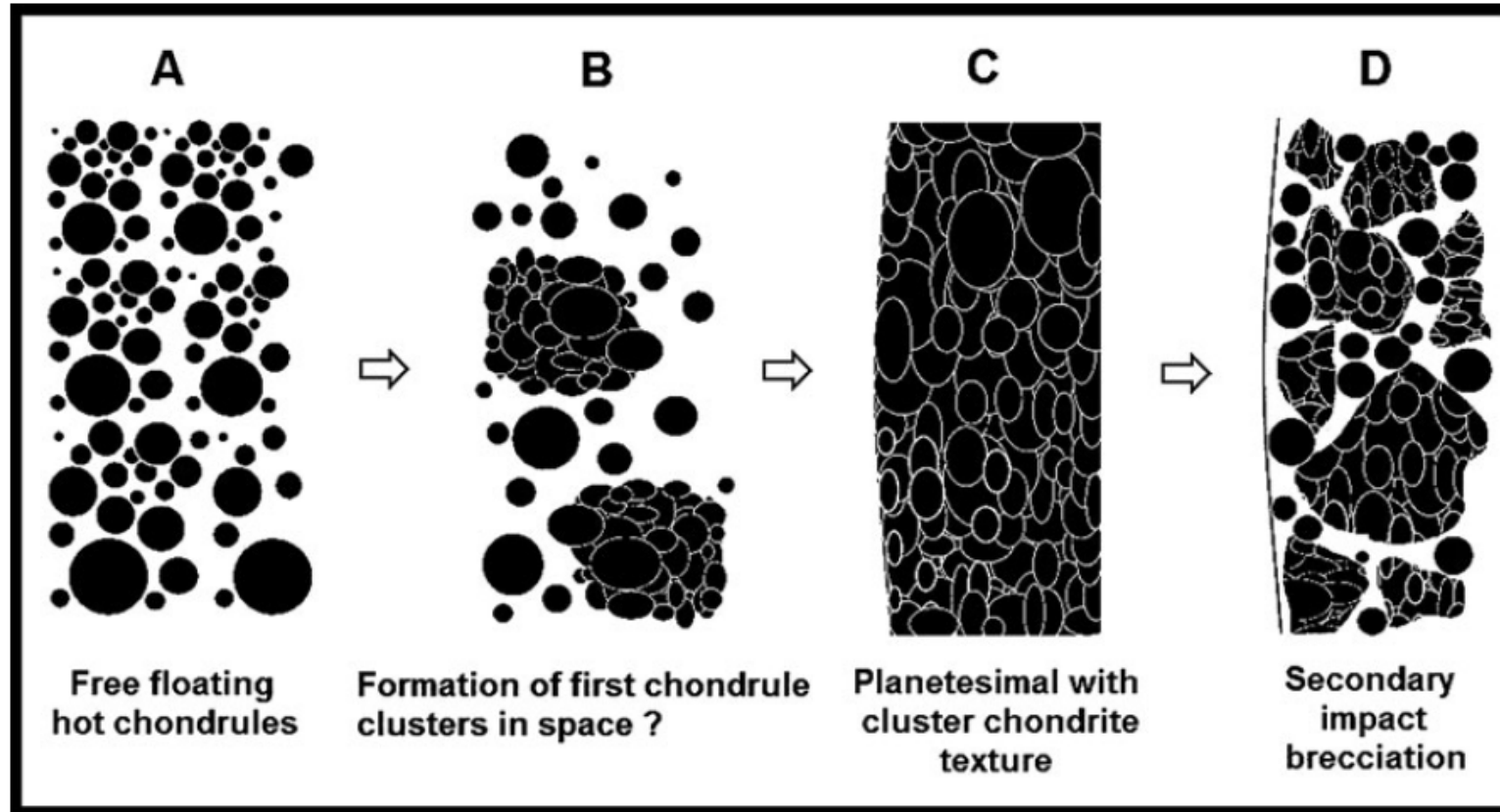
(Taillifet, Charnoz, Aléon & Baillie, in prep)

**Krymka chondrite
(LL3.2)**

When did the accretion take place?

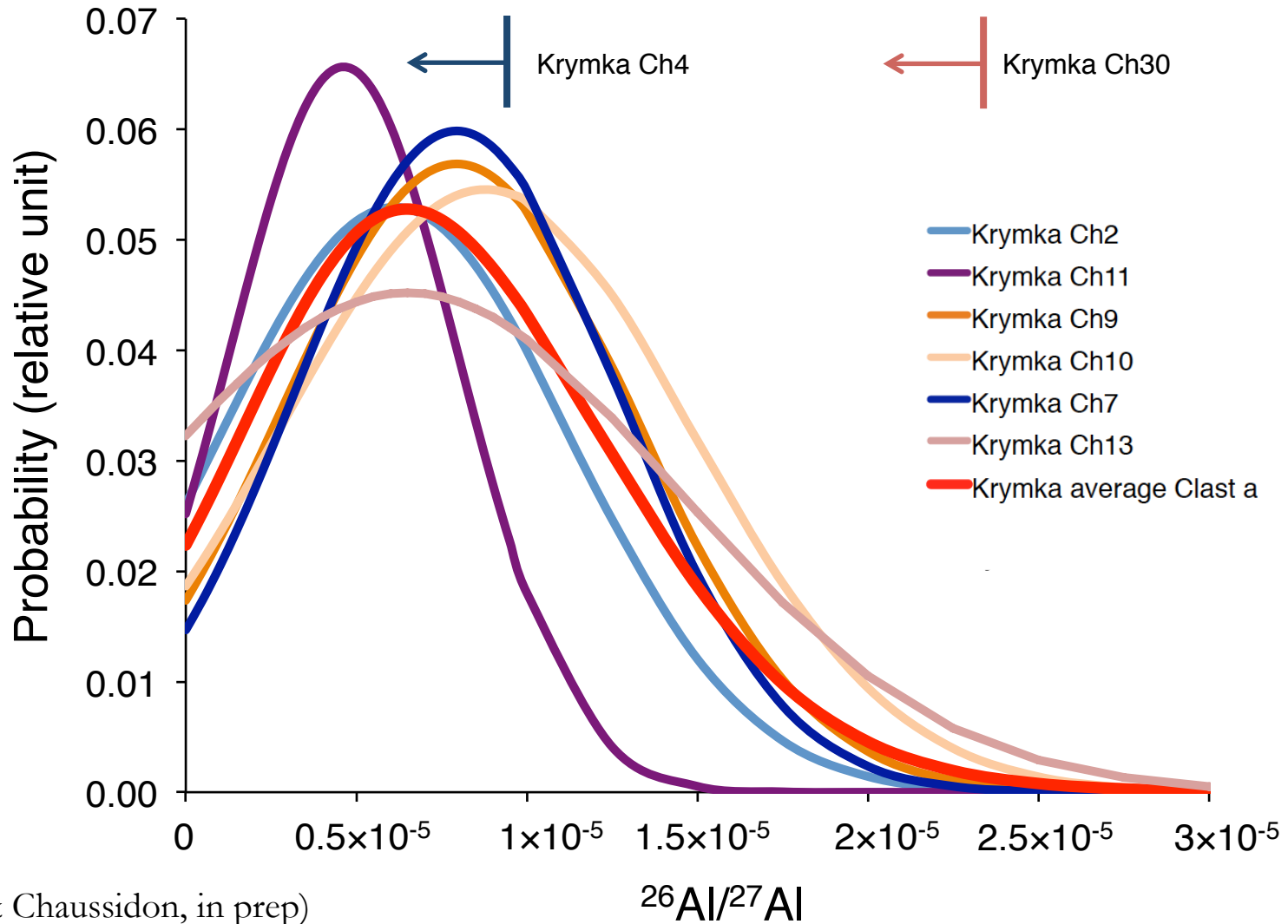


Accretion of hot chondrules (Metzler, 2012)



If the chondrules are deformed because they accreted being still hot, then the ^{26}Al age given by their mineral isochrons should all be the same and date the accretion

All chondrules in clast a are in agreement with having been quenched below the closure temperature of Mg isotopes when $^{26}\text{Al}/^{27}\text{Al}=6.9(\pm 5.5)\times 10^{-6}$,
(i.e. $2.1_{-0.6}^{+1.7}$ Ma after CAIs)



(Metzler & Chaussidon, in prep)

October 26th to 31st

Labex **UnivEarthS**

FALL SCHOOL 2014

Villa Finaly, Florence (Italy)



Clues from the chemical and isotopic composition of the Earth
on its origin in the Solar protoplanetary disk

CONCLUDING REMARKS

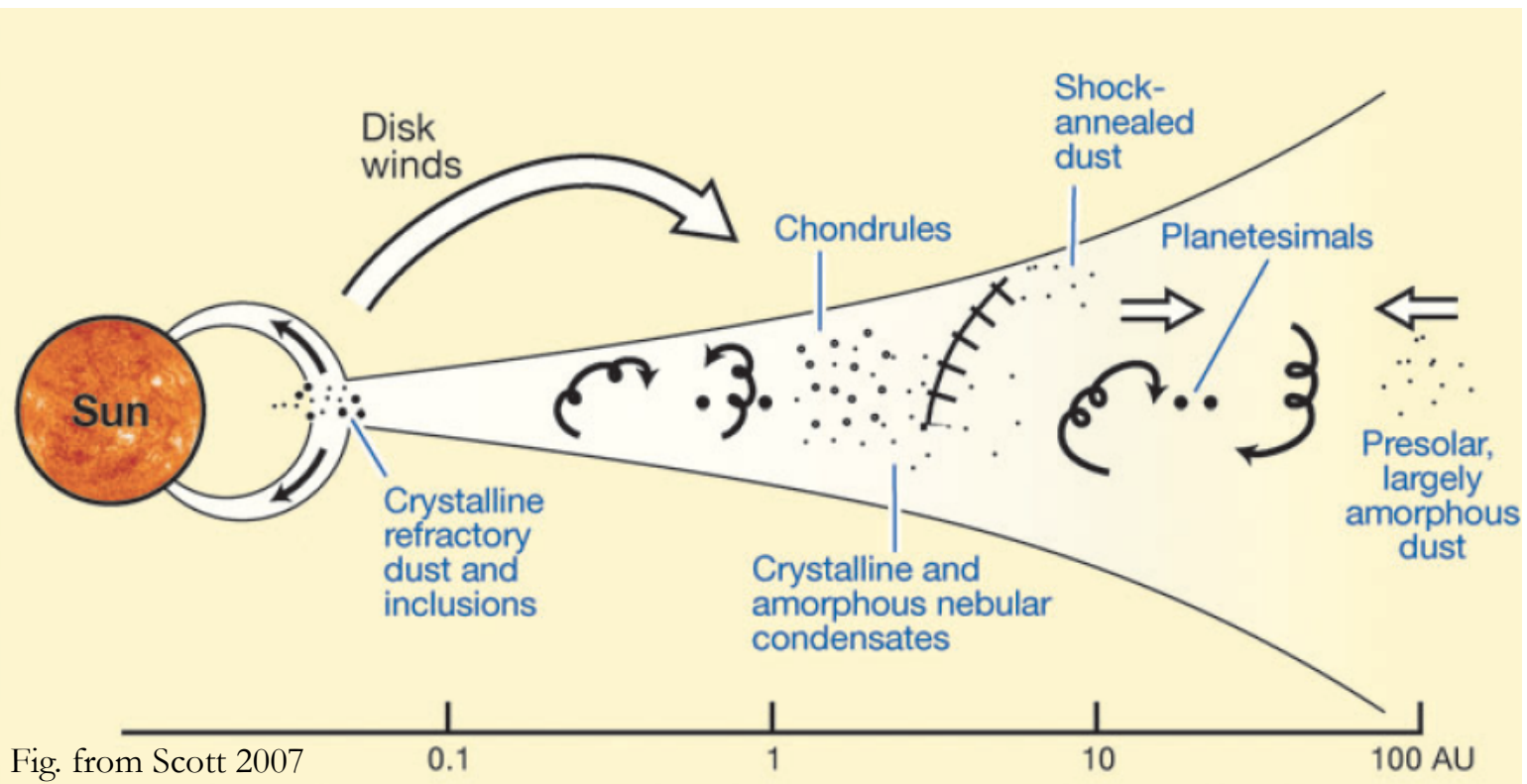
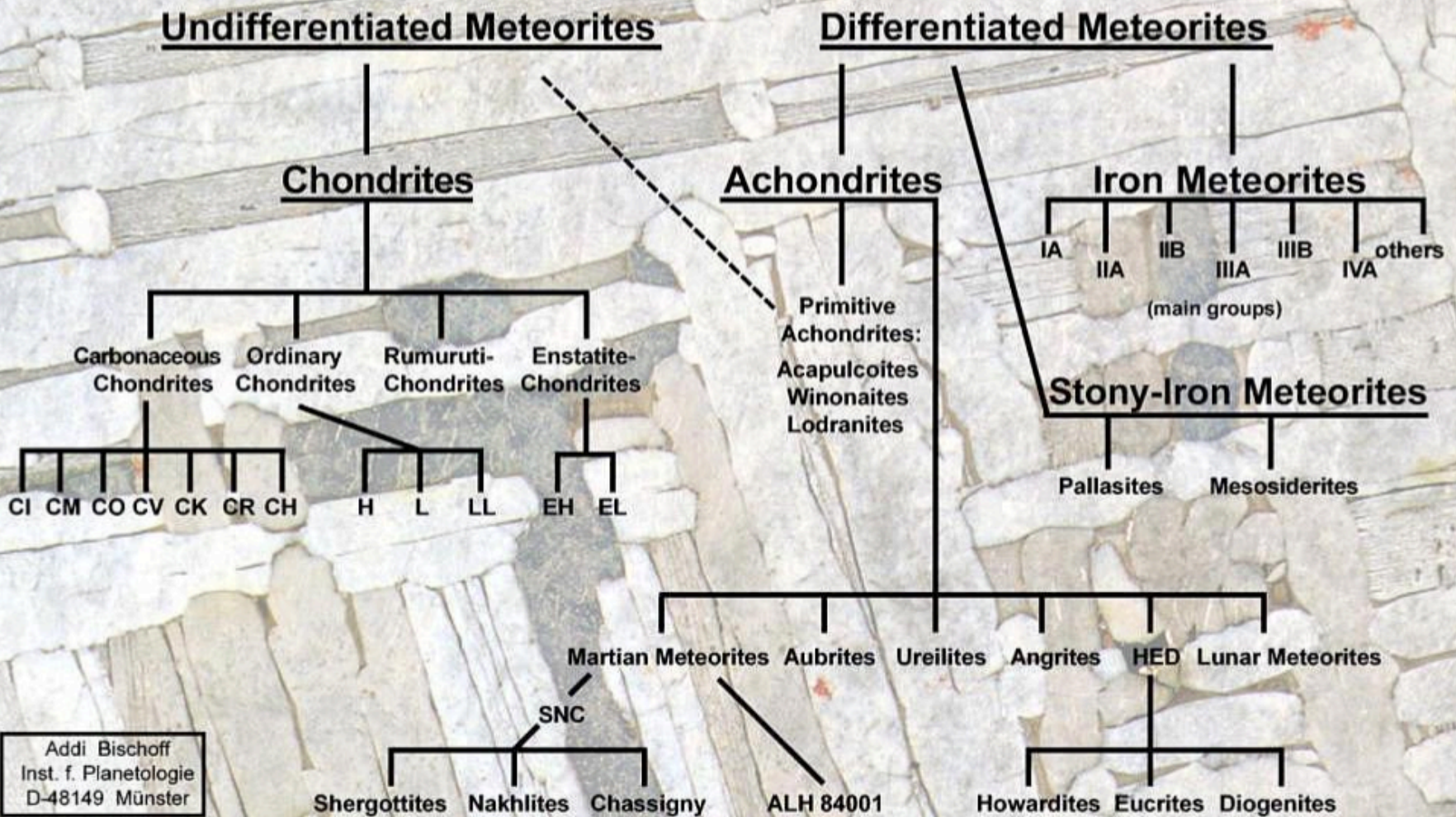


Fig. from Scott 2007

COSMOS
MODELING

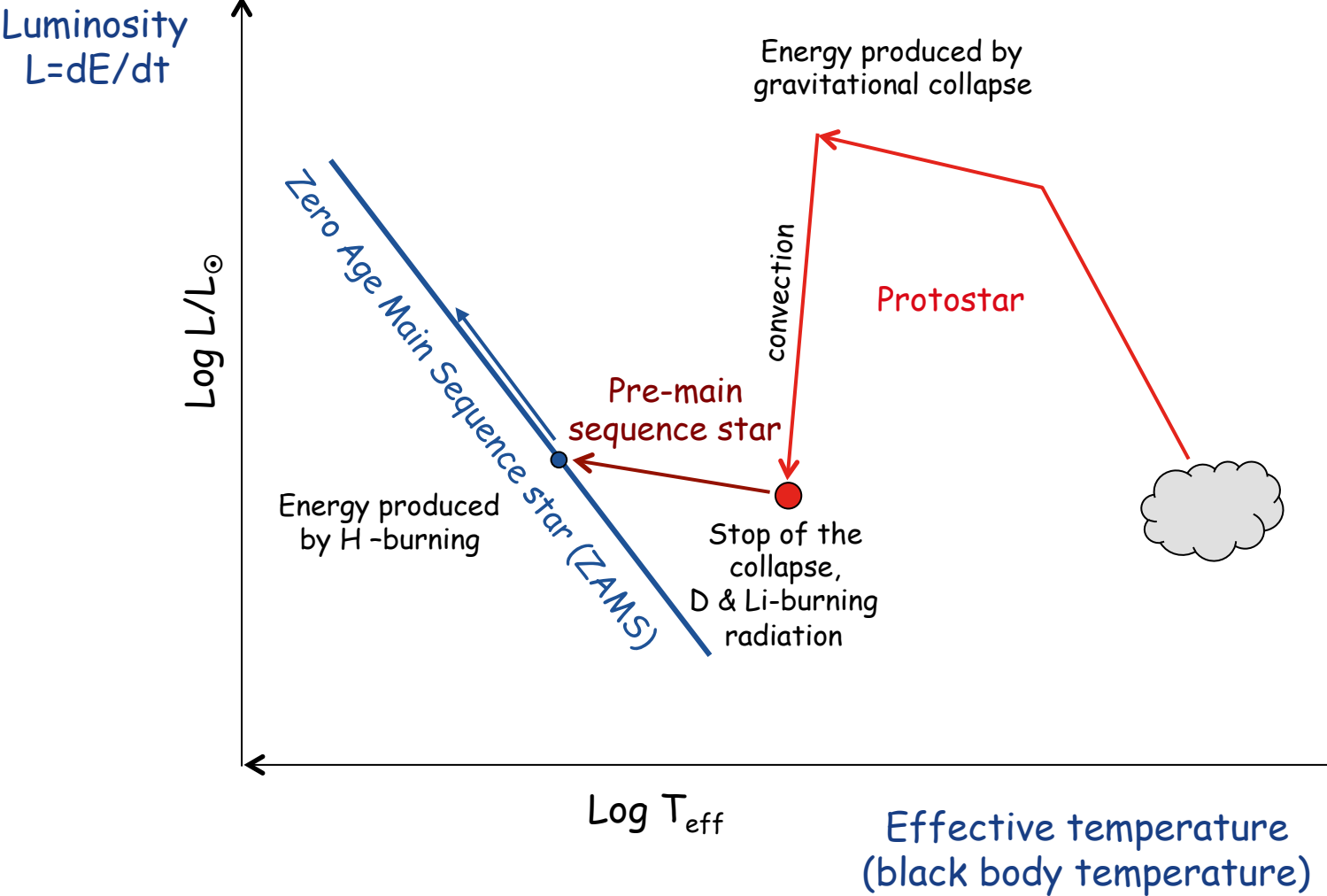


Meteorite Classification

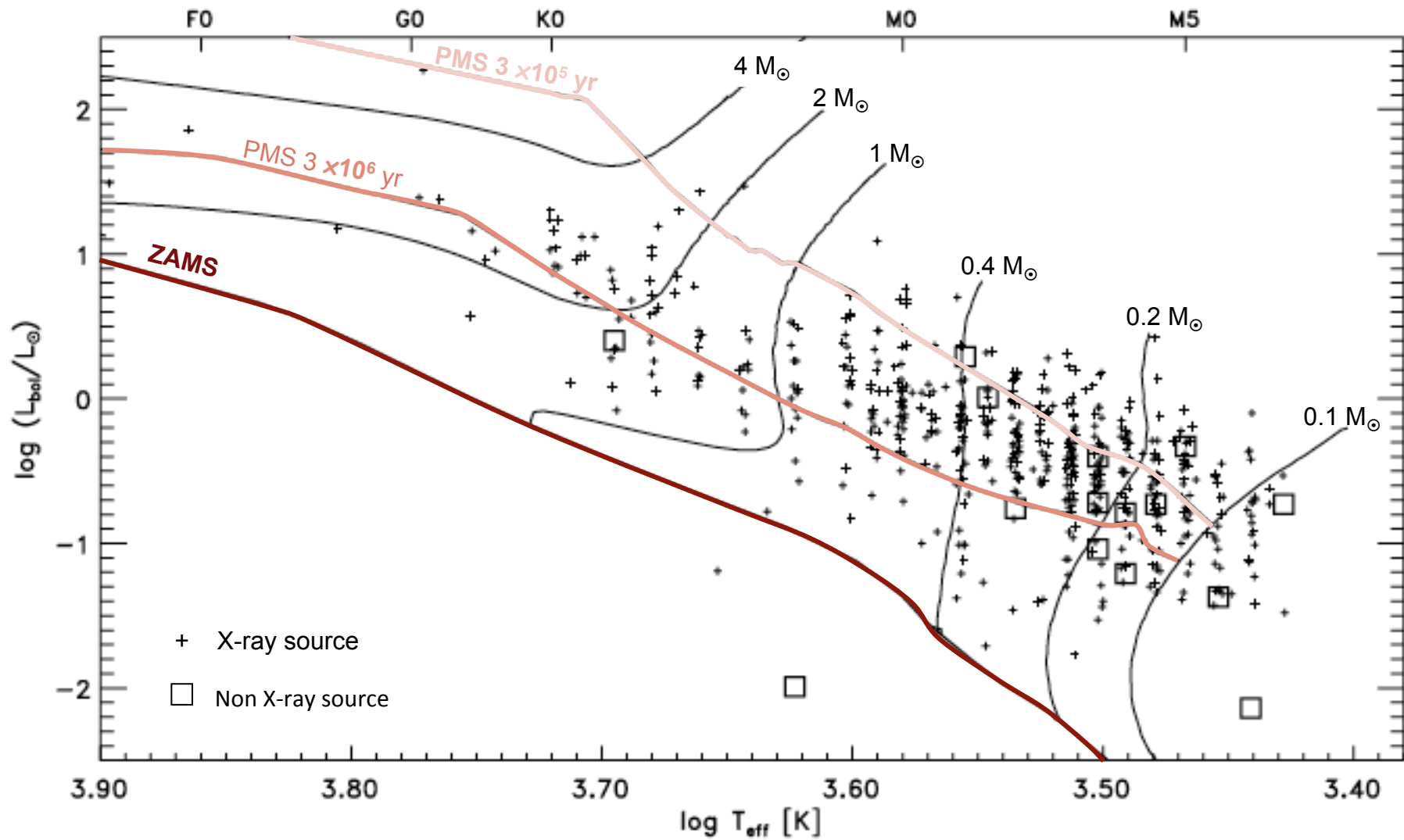


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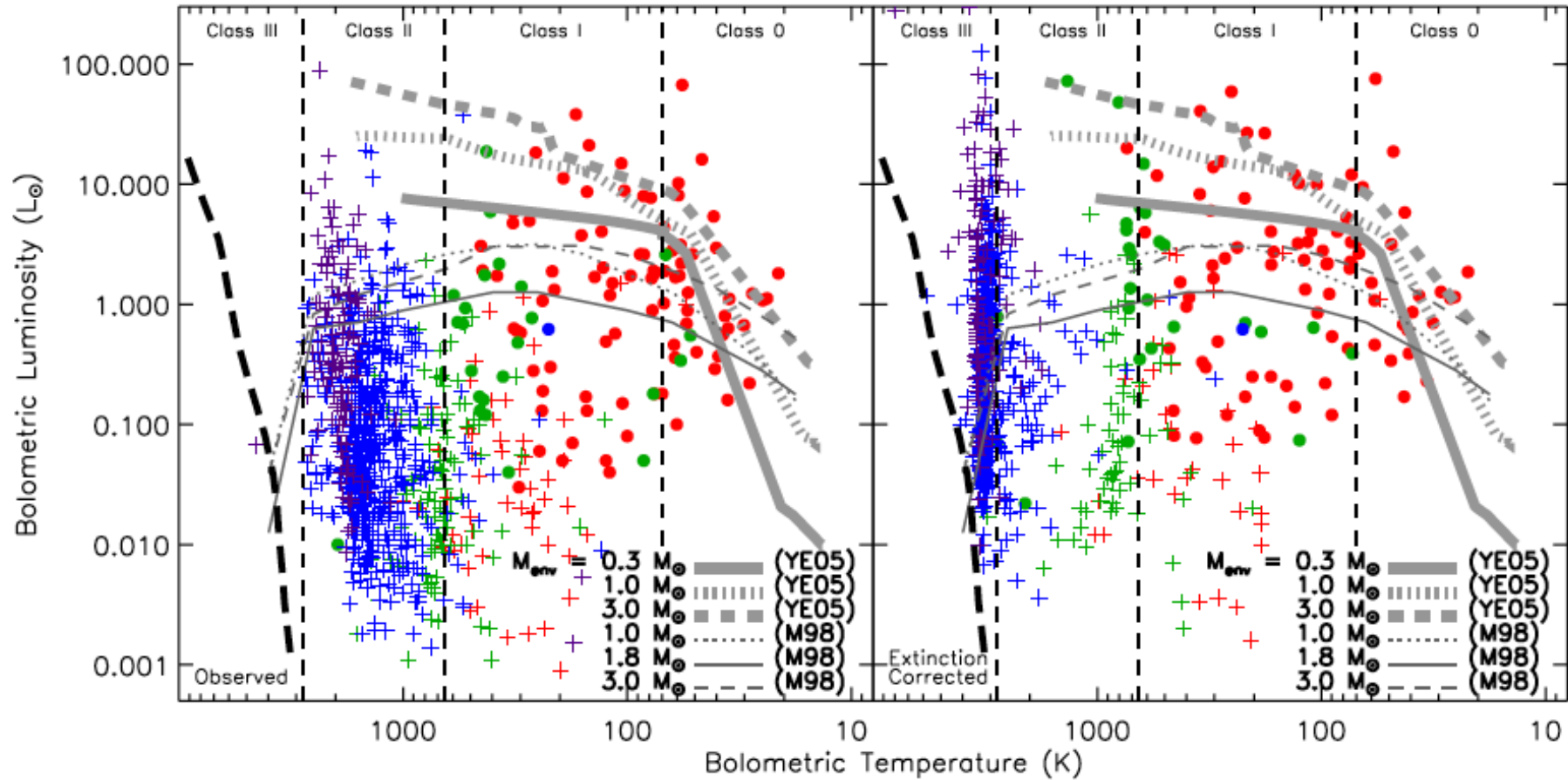
Schematic view of the Hertzsprung-Russel diagram for protostars evolving towards the main sequence



Pre-main sequence stars identified by their intense X-ray emissivity



Statistic observations of young stars (>1000 objects in 5 clouds)



Evans et al., 2009

Classification of Lada (1991) based on spectral index

Dots=sources with envelopes still present

- Protostar: Class I (red), Flat (green)
- Pre-main sequence: Class II (blue), Class III (purple)