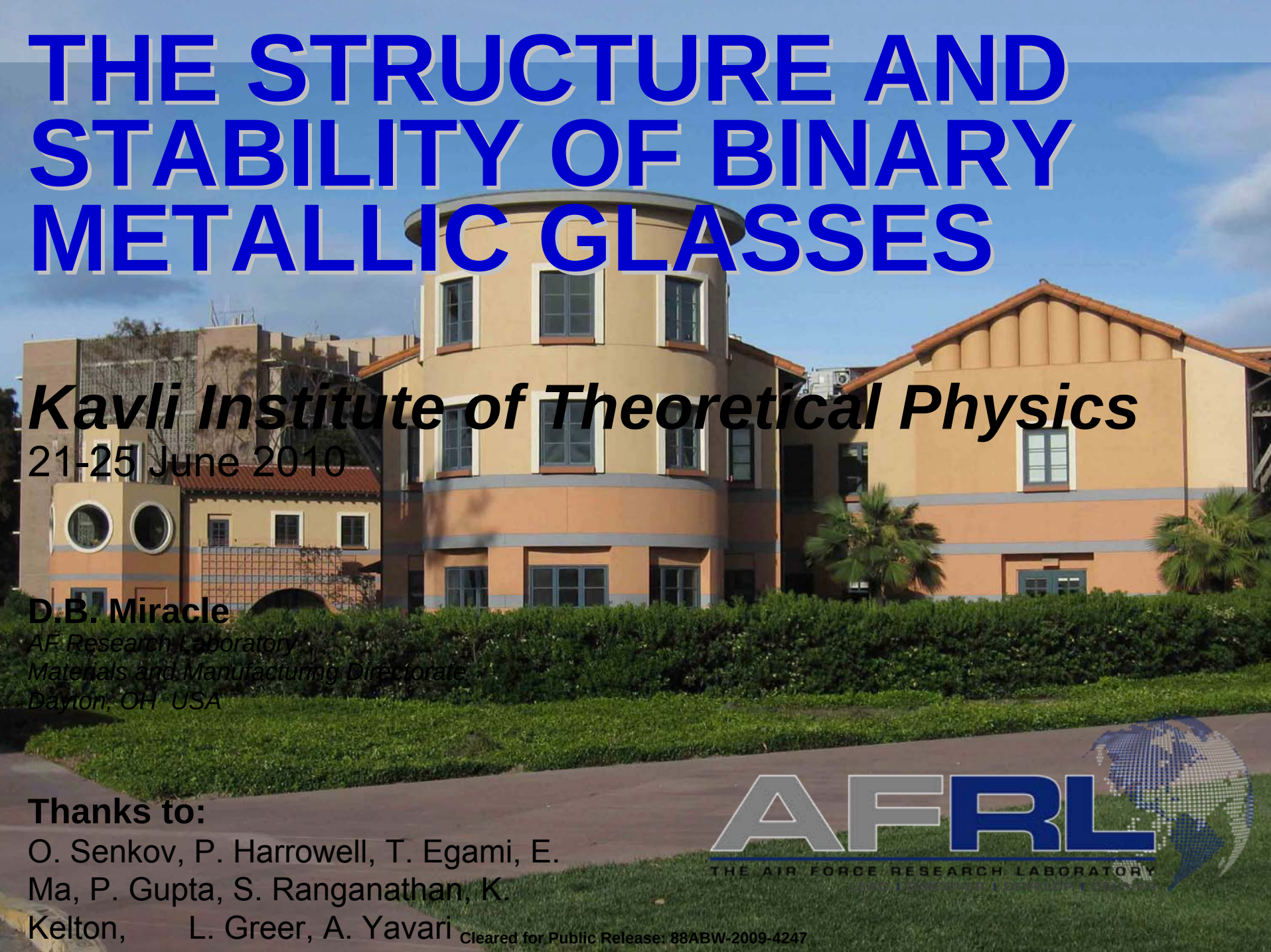
THE STRUCTURE AND STABILITY OF BINARY METALLIC GLASSES



Kavli Institute of Theoretical Physics

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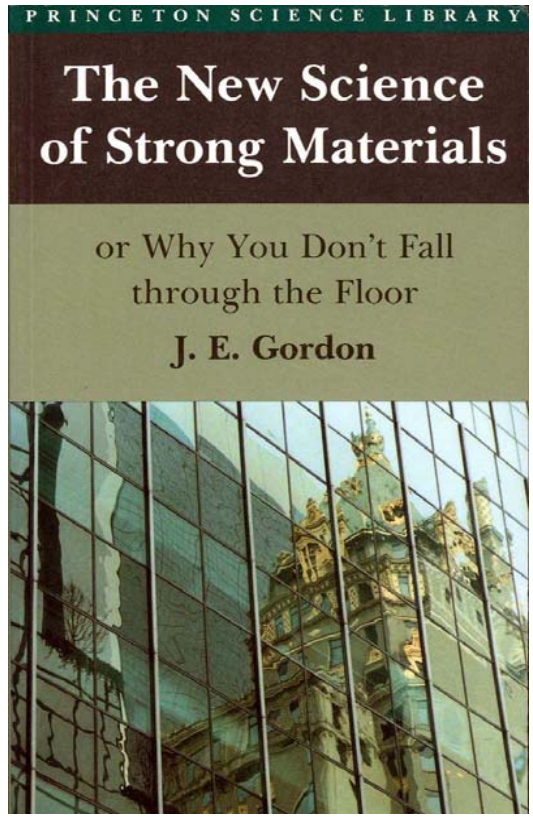
Thanks to:

O. Senkov, P. Harrowell, T. Egami, E. Ma, P. Gupta, S. Ranganathan, K. Kelton, L. Greer, A. Yavari

experimentalists

"~~metallurgists~~ ^ are apt to be practical down-to-earth people who stand no nonsense, but the theoriticians

~~non-metallurgists~~ ^ are probably more lyrical and imaginative"



EARLIER INSIGHTS

“This, in turn, suggests correlated rather than random arrangements of local structural units.” (P.H. Gaskell, 1983)

“We can no longer assume, I believe, that we can think in the seductive simplicity of the language of randomness alone.” (P.H. Gaskell, 1991)

“. . . the amorphous state is in reality not a disordered state, but a rather well organized arrangement of atoms. . .” (S. Steeb and P. Lamparter, 1993)

“But more recent work has shifted the balance of evidence towards structures that are more complicated, more diverse and more ordered– at least in the sense that there may be an underlying ordering or structure-forming principle.” (P.H. Gaskell, 1991)

“My own view is that simple geometry. . . atomic sizes. . . will prove to be the main criterion that in various subtle ways incorporates the others.” (R.W. Cahn, 1991)

SUMMARY

Show that metallic glasses are distinguished by:

- Atomic packing that is efficient over local and global length scales
- Preferred relative atomic sizes
- Specific short-range and medium-range (~1 nm) atomic order
- Binary, ternary or quaternary atomic size distributions
- Solute concentrations with a minimum of 5-10% and preferred >30%

Give a simple, 3D portrait of binary metallic glass structures

- Physically-based and experimentally-validated description of SRO, MRO

Show that only a small subset of topologies (relative size and concentration) are common, and a smaller subset are relatively stable

- All binary BMGs have $R \cong 0.71$, 0.80, 1.12 or 1.25 and are solute-rich

Introduce importance of relative inter-atomic bond enthalpies

EYESIGHT *vs* INSIGHT
Why not measure structure directly?

Early investigations used 'eyesight'

- State-of-the-art experimental methods nevertheless lose important details
- Low spatial resolution provides globally averaged signals
- Reliable information primarily available for first few atomic coordinations
- Deconvolution of overlapping signals is required to quantify atomic coordinations and separations
- Large integration limits produce non-unique results

Direct experiment gives only broad statistical descriptions

Computations provide an alternate approach to describe structure

- Individual simulations are ultimately system-specific
- The information produced is overwhelming, so that a system for organizing results is needed

METALLIC GLASS FEATURES

What is known already?

A credible structural model must show agreement with established metallic glass characteristics:

- randomness is a dominant and defining feature
- efficient atomic packing is required over all length scales
  - *low molar volume, small density decrease upon solidification and crystallization and quantification of locally efficient atomic packing are all observed*
- strong short range ordering (compositional and topological)
- significant medium range order (compositional and topological)
- significant size difference (≥12%) between solvent and solute atoms
- large negative enthalpy of mixing of constituent elements
- three or more solutes
- relative sensitivity in some glasses to small composition changes (~1%)
- relative insensitivity in some glasses to large composition changes (~10%)

A compelling structural model will give a predictive capability for many of these features

HISTORY OF EFFICIENT PACKING

Kepler Conjecture is an intuitive solution to a 'simple' problem

David Hilbert highlighted efficient packing in a list of problems to guide mathematics in the 20th century

- "How can one arrange most densely in space an infinite number of equal solids of given form?"

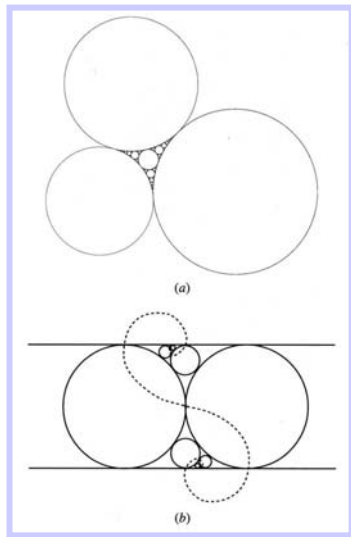
Mathematics has extended intuition and experience

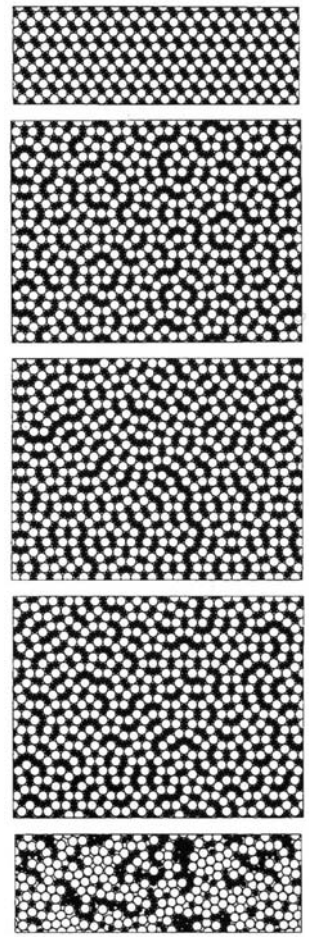
- Solution to Kepler Conjecture claimed in 1998

Significant additional complexity exists in systems of *unequal spheres*

- the number and relative sizes of spheres becomes important
- binary or complex size distributions may exist
- relevant in problems from concrete to cosmology

Figure 3.8. Johannes Kepler (1571–1630).





OUTLINE

LOCAL STRUCTURE
Efficiently-packed solute-centered clusters

EXTENDED STRUCTURE

BINARY METALLIC GLASS STRUCTURES

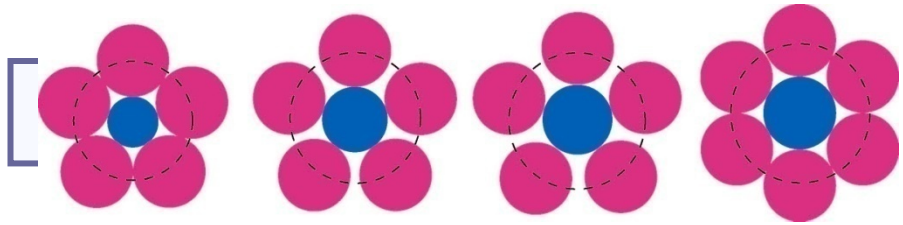
BINARY METALLIC GLASS ASSESSMENT

SUMMARY

EFFICIENT LOCAL PACKING
2D

The 2D theoretical coordination number (N^T) is the number of circles of radius r_j that can be placed around a central circle of radius r_i, where $R = r_i/r_j$.

- N^T is a real number



$$N^T = \pi/\arcsin[1/(1+R)]$$

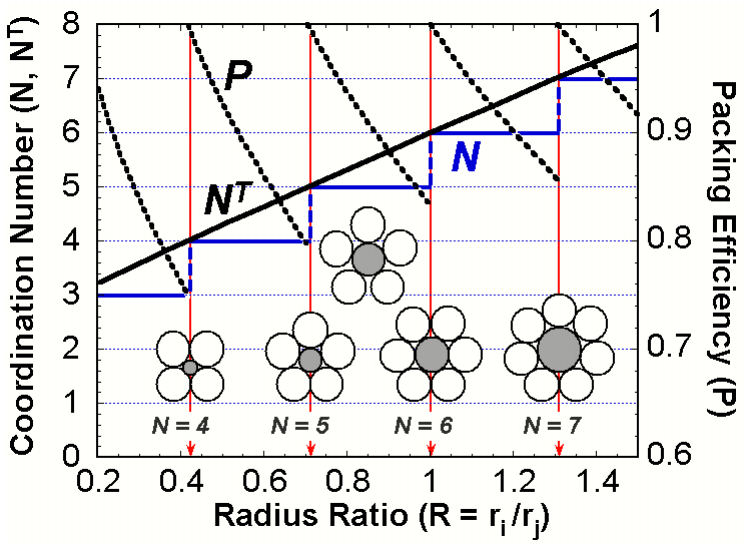
Egami and Waseda; *J. Non-Cryst. Sol.*, **64**, 113–134(1984)

Packing Efficiency (P) is the maximum number of full circles of radius r_j that can be placed around a central circle of radius r_i, normalized by N^T

$$P = Trunc(N^T)/ N^T$$

Packing efficiency varies with R

- P is highest when the first shell is completely 'filled' with no gaps
- P is highest for specific values of R where N^T is an integer (R^*)

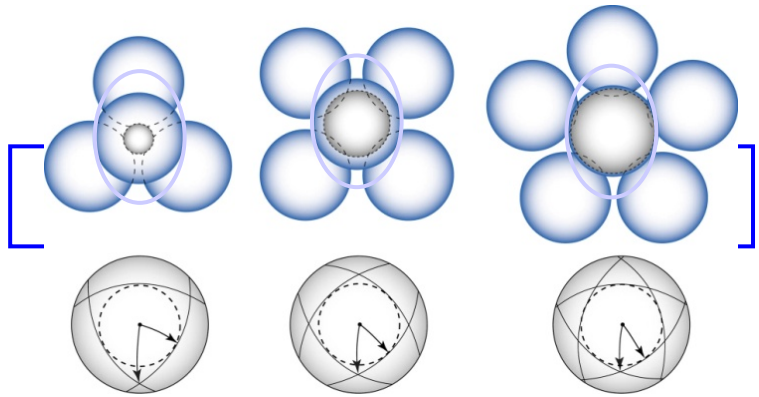


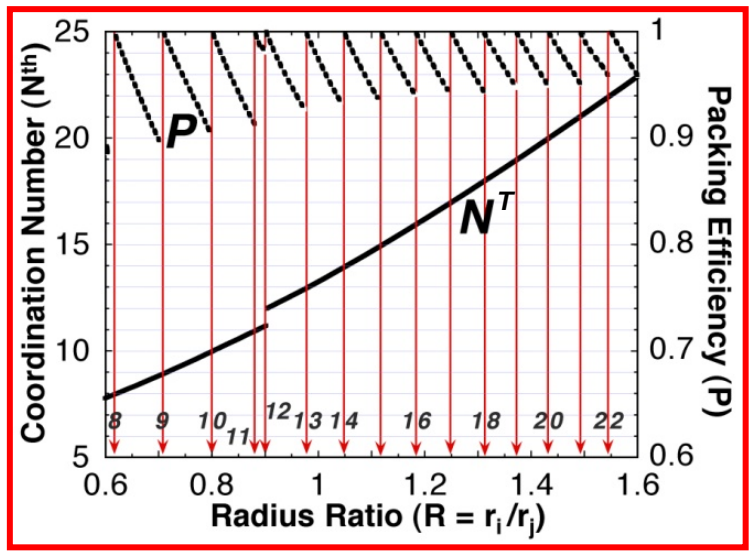
EFFICIENT LOCAL ATOMIC PACKING (ELAP)

3D relationship given between *R* and packing efficiency in 1st atomic shell

- Packing efficiency is a maximum when N^T is an integer
- N^T is an integer for specific ratios, R^*
- Suggests that specific radius ratios R^* may be preferred in metallic glasses

N^T	R^*	N^T	R^*
6	0.414	**14**	1.047
7	0.515	**15**	1.116
8	0.617	**16**	1.183
9	0.710	**17**	1.248
10	0.799	**18**	1.311
11	0.884	**19**	1.373
12	0.902	**20**	1.433
13	0.976	**21**	1.491





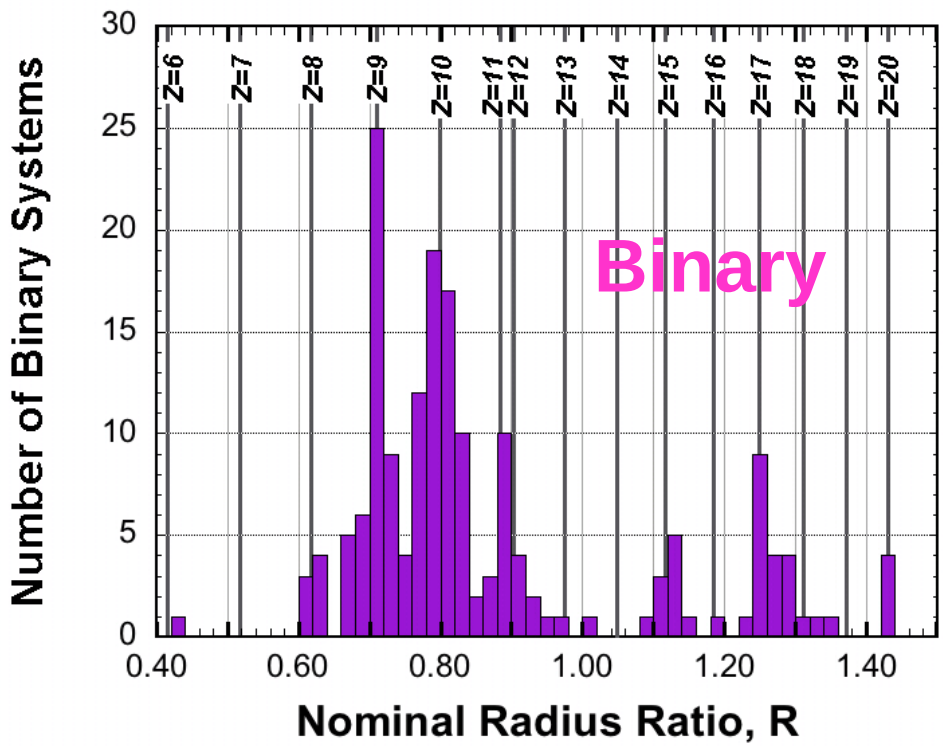
Miracle, Sanders and Senkov; *Phil Mag. A*, **83**, (2003)

PRINCIPLE 1

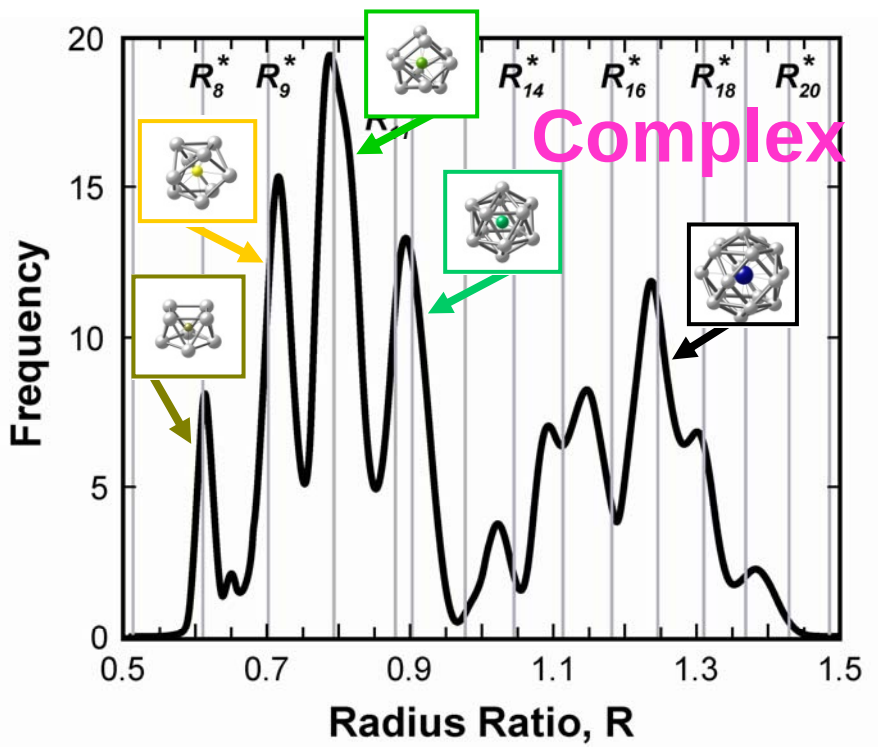
*ELAP Via Preferred Relative Atom Sizes, R**

Analysis of >400 radius ratios in metallic glasses confirms strong preference for R^* values

– Efficient local atomic packing is concluded to be important in the formation of metallic glasses



Miracle, Louzguine, Louzguina, Inoue; *Int'l Mater. Rev.*, In Press.



Miracle, Sanders and Senkov; *Phil. Mag.*, **83A**, (2003)

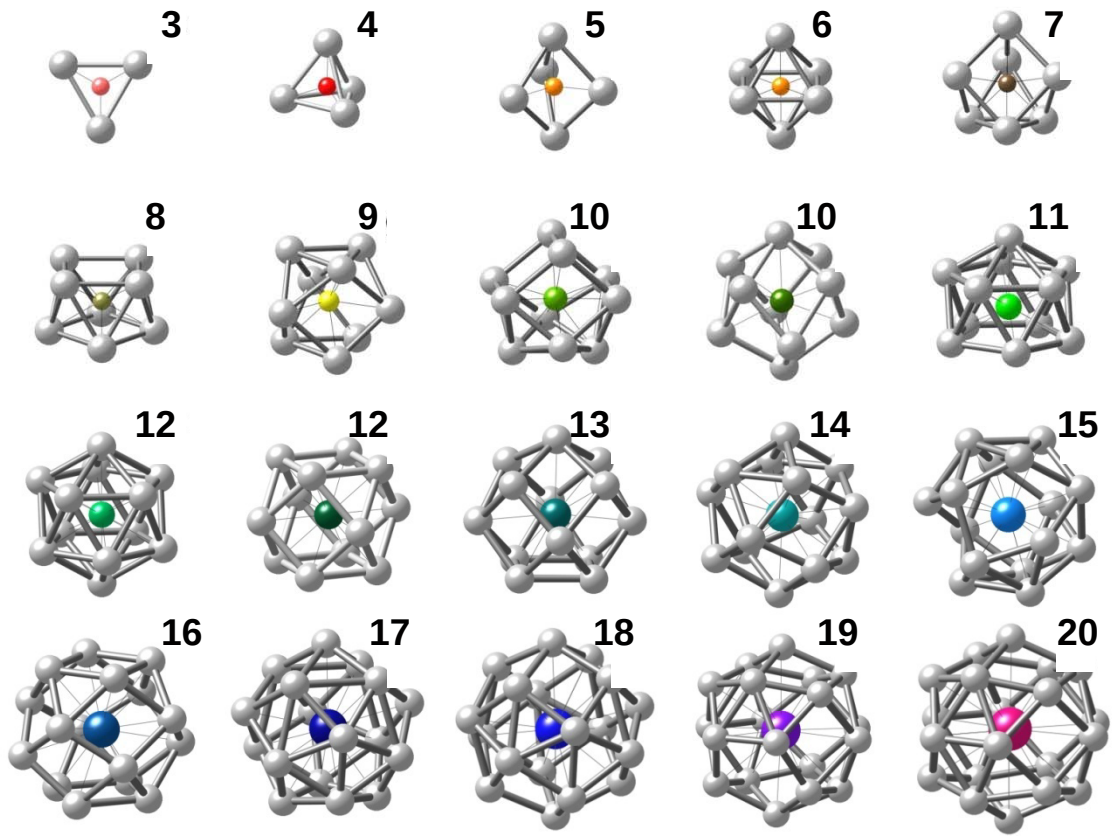
EFFICIENT LOCAL ATOMIC PACKING (ELAP)

Changing solute-to-solvent radius ratio enables efficient atomic packing in the 1st coordination shell

- Specific solute-to-solvent radius ratios are preferred

Efficiently packed solute-centered canonical clusters with specific *N, R*

- Can be considered as local representative structural elements
- Each *N* introduces a family of clusters
- Many different local clusters



Miracle, Lord and Ranganathan; *Trans. JIM*, **47**, 1737 (2006)

OUTLINE

LOCAL STRUCTURE

EXTENDED STRUCTURE
Filling of Space by Efficiently-Packed Clusters

BINARY METALLIC GLASS STRUCTURES

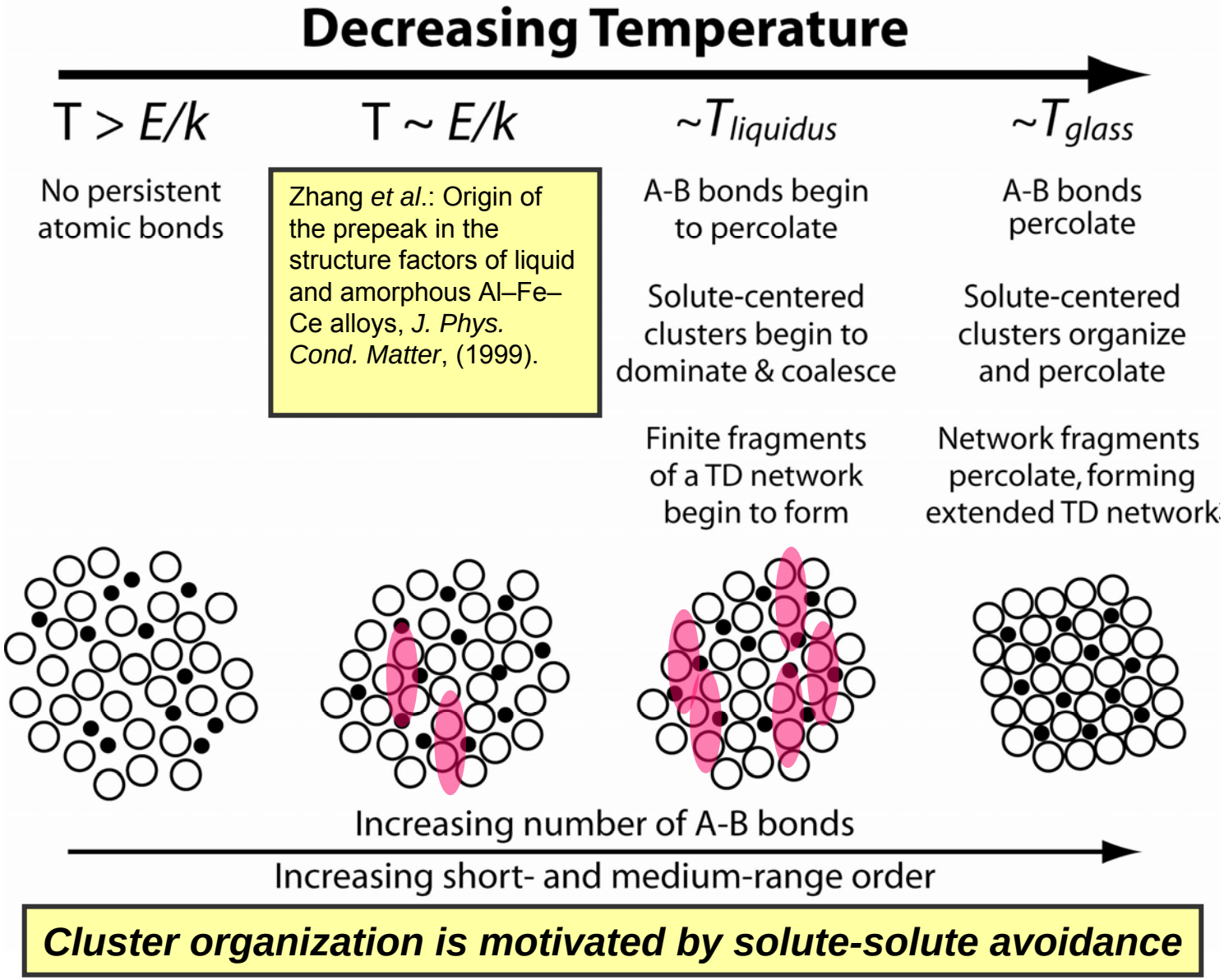
BINARY METALLIC GLASS ASSESSMENT

SUMMARY

Miracle, *Nature Mat.*, **3**, 697 (2004);
Miracle, *Acta mater.*, **54**, 4317 (2006)

CLUSTER ORGANIZATION

Evolution of MRO from the Liquid



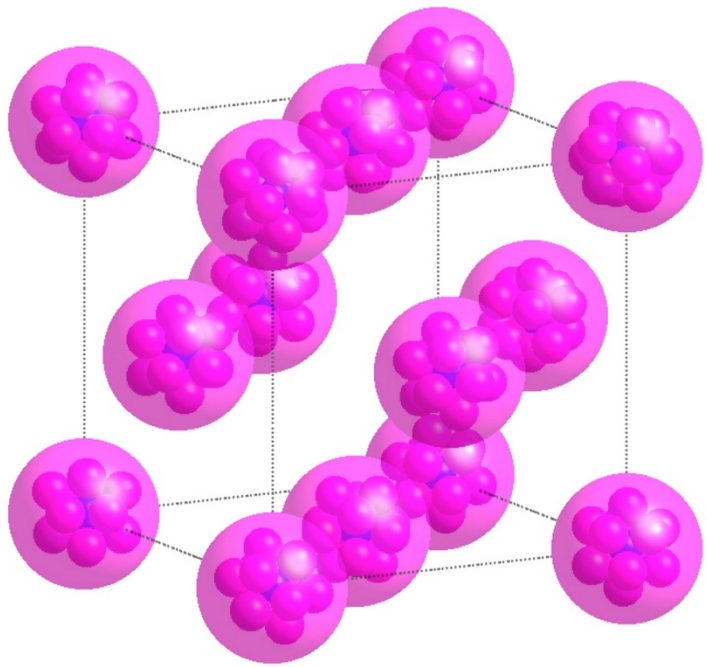
Cluster organization is motivated by solute-solute avoidance

PRINCIPLE 2
Efficient Packing of Primary Clusters

Efficiently-packed solute-centered atomic clusters are imagined to be sphere-like

Efficient atom packing beyond the 1st atomic shell is achieved by dense packing of these sphere-like clusters

- fcc, bcc, hcp, sc, icosahedral and random cluster packing considered
- fcc cluster packing gives the most efficient packing of equal-sized spheres and best agreement with measured MRO



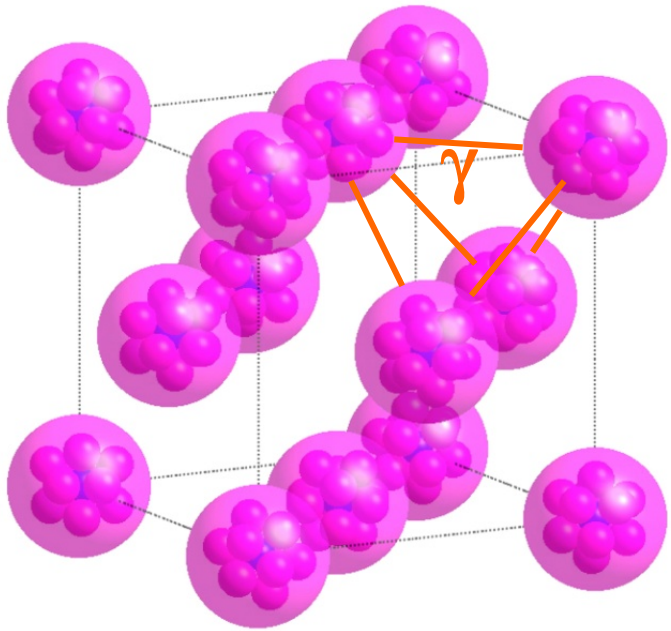
Efficiently-packed, solute-centered clusters are organized in space to achieve efficient cluster packing

PRINCIPLE 2 CONSEQUENCES

Efficient Packing of Primary Clusters

Four topologically distinct atomic species and sites

- Solvent atoms (Ω)
- Primary (α) solutes produce the structure-forming unit clusters
- Cluster-octahedral interstices (β)
- Cluster-tetrahedral interstices (γ)
- $r_\alpha > r_\beta > r_\gamma$



Solute atoms occupy ~ordered sites

- Provides basis for observed medium range atomic ordering (MRO)
- Variable cluster-cluster separation degrades cluster ordering beyond a few cluster diameters

Preferred atom positions introduces the possibility of structural defects

- Vacancy and anti-site point defects
- Constitutional and thermal

“Ninety-nine percent loyalty is 100% disloyalty.”

Napoleon Bonaparte

“He’s not dead. I said he’s *mostly* dead. BIG difference.”

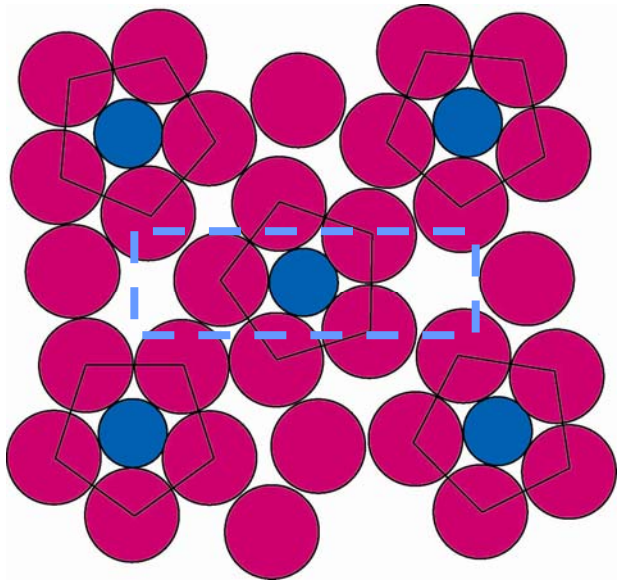
Miracle Max, from “The Princess Bride”

“The Pirate’s Code is more what you’d call ‘guidelines’ than actual rules.”

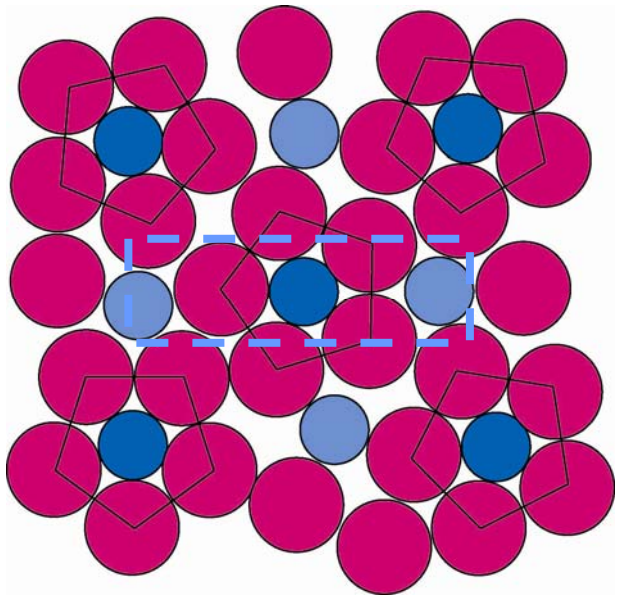
Captain Barbossa, from “The Pirates of the Caribbean”

STRUCTURE AND BONDING

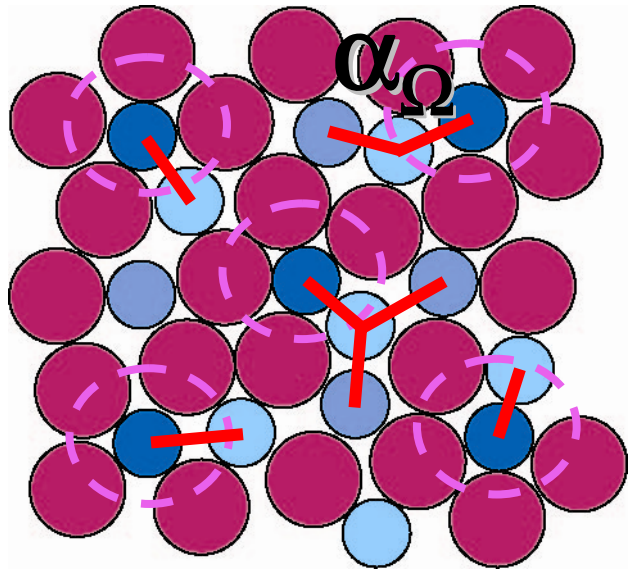
α–α Bonds in Solute-Rich Glasses



β vacancies (V_β)



α anti-site defects on β sites (α_β)



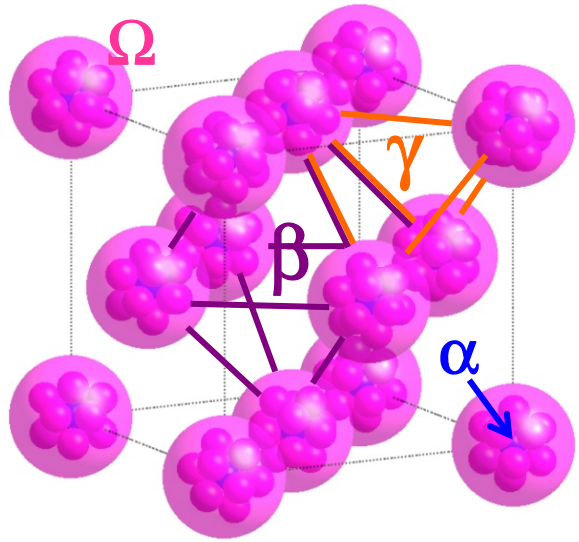
α anti-site defects on Ω sites (α_Ω)

EFFICIENT CLUSTER PACKING

Principles of Construction

Solutes have specific radius ratios, *R

– R_Z^* enables efficient local atomic packing of Z solvent atoms (Ω) around a solute atom (α)

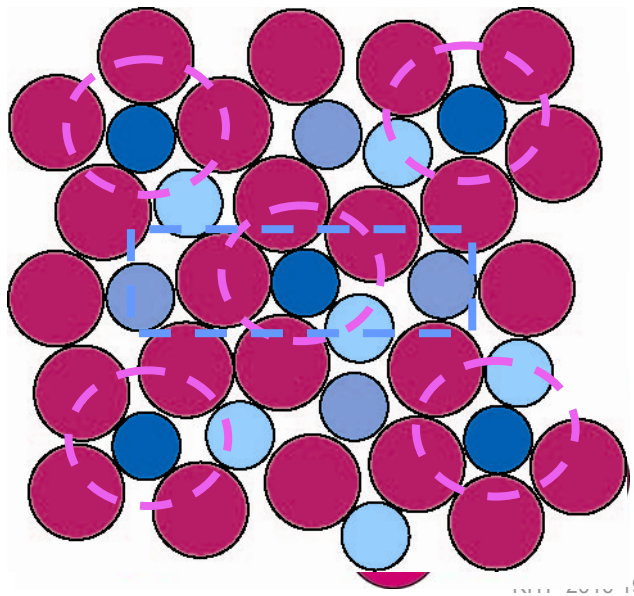


Efficiently-packed, sphere-like, solute-centered clusters are organized in space to achieve efficient cluster packing

– ~Cubic close-packing of clusters enables efficient global atomic packing

➢ *Gives quantitative agreement with diffraction data*

– Solvent sites and 3 solute sites are produced



No orientational order between clusters

Preferred site occupancies

– Solute atoms occupy solute sites (α, β, γ)

– Solute atoms occupy Ω sites (α_Ω defects)

Supported by atomistic simulations

Sheng, Luo, Alamgir, Bai, Ma: *Nature*, **439**, 419 (2006).

STRUCTURAL TOPOLOGIES

A wide range of glass topologies (276) are possible

BINARY	TERNARY	QUATERNARY
<8>		
<9>	<9,8>	
<10>	<10,9>, <10,8>	<10,9,8>
<11>	<11,10>, <11,9>, <11,8>	<11,10,9>, <11,10,8>
<12>	<12,11>, <12,10>…	<12,11,10>, <12,11,9>…
<13>	<13,12>, <13,11> …	<13,12,11>, <13,12,10> …
<14>	<14,13>, <14,12> …	<14,13,12>, <14,13,11> …
<15>	<15,14>, <15,13> …	<15,14,13>, <15,14,2> …
<16>	<16,15>, <16,14> …	<16,15,14>, <16,15,13> …
<17>		
<18>		
<19>		
<20>		

<12-10-9> Glass

<17-12-10> Glass

Do all topologies have the same stability, or are some intrinsically more stable?

Topological stability favored when:

- Solute sites are filled by solutes that fit most naturally
- The number of unlike atomic bonds are maximized

OUTLINE

LOCAL STRUCTURE

EXTENDED STRUCTURES

BINARY METALLIC GLASS STRUCTURE

Structural Site Occupancies

Model Validation

BINARY METALLIC GLASS ASSESSMENT

SUMMARY

Miracle, *Nature Mat.*, **3**, 697 (2004)

Miracle, *Acta mater.*, **54**, 4317 (2006)

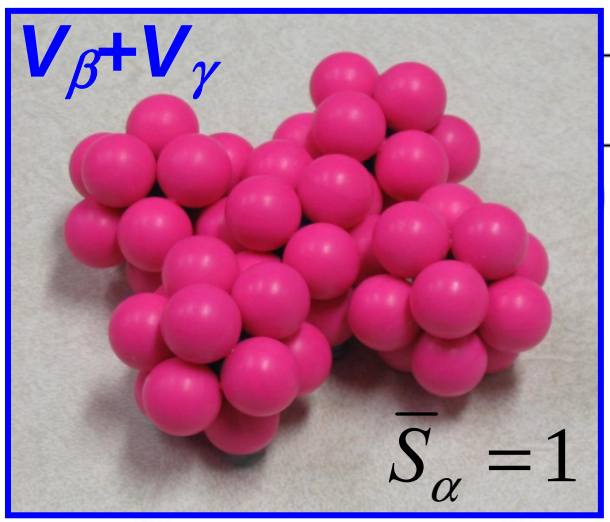
Miracle, Louzguine, Louzguina, Inoue; *Inter. Mater. Review*, In press.

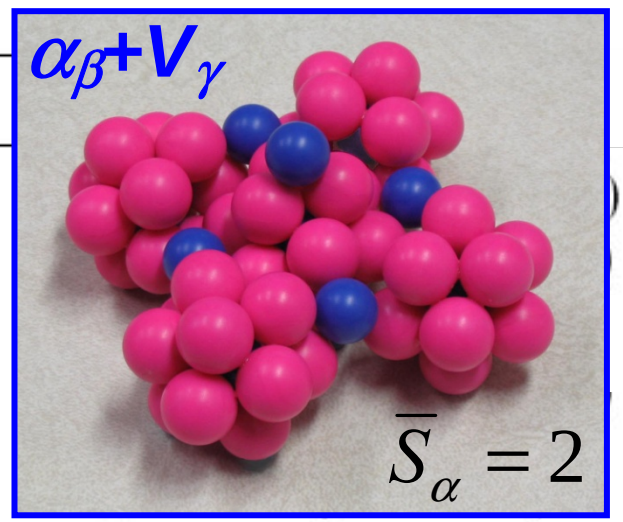
STRUCTURAL CHARACTERIZATION

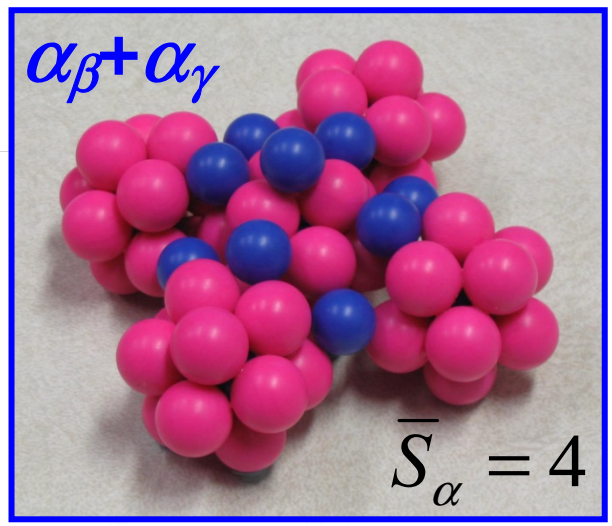
Site occupancies $S(i_j)$ are the primary structural descriptors

- $S(i_j)$ is the number of species, *i*, that occupy sites, *j*, per α site
- The total number and types of sites are known
- $S(i_j)$ is obtained by matching composition
  - *Fill solute sites in the order* α, β, γ, Ω

$$\bar{S}_\alpha = \left(F_\alpha / F_\Omega\right)\left(\hat{S}_\Omega - S(\alpha_\Omega)\right)$$







BINARY GLASS TOPOLOGIES

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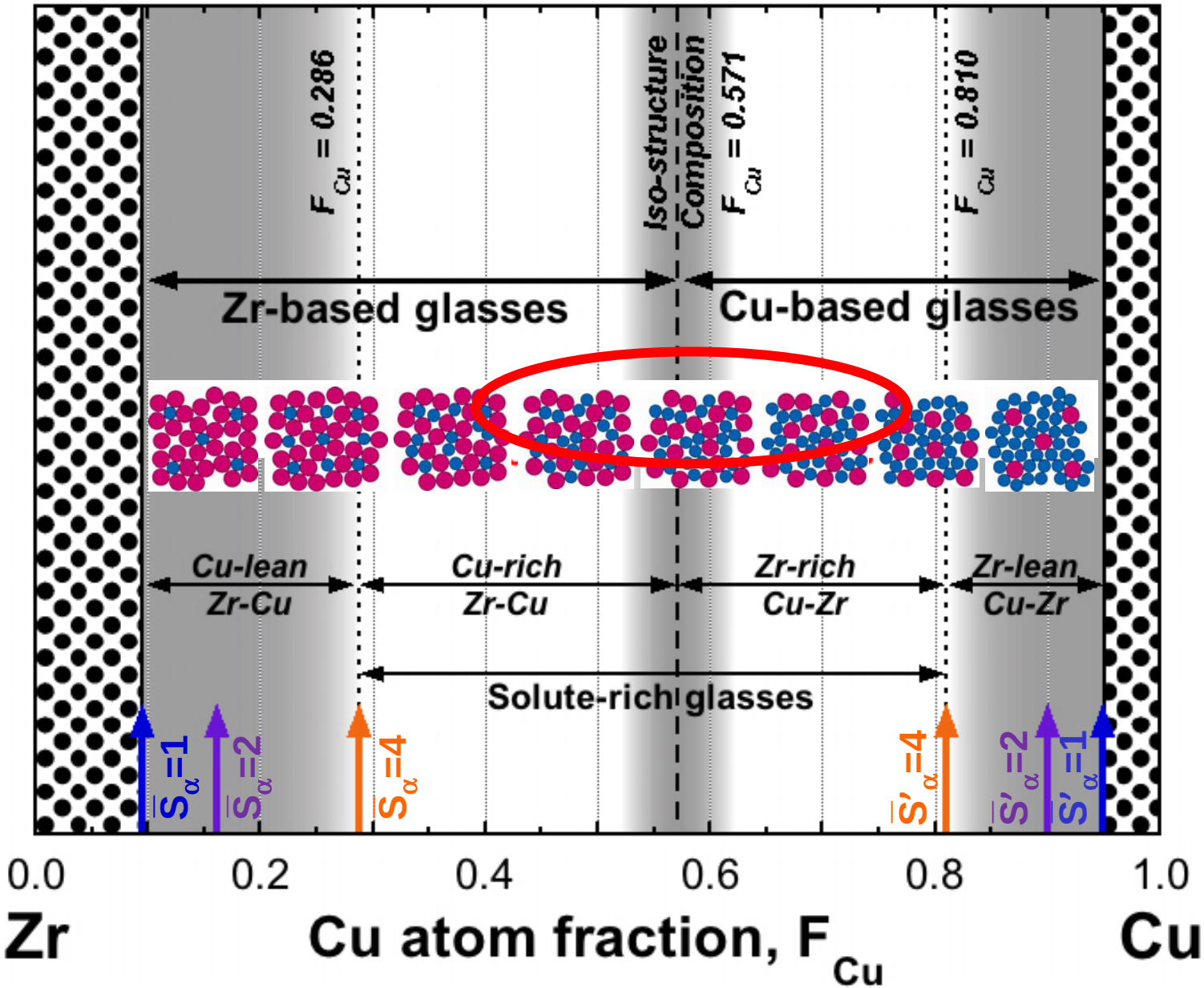


SHORT RANGE ORDER
α Nearest-Neighbor Coordinations

artial coordination of α around α ($N_{\alpha-\alpha}$)

$$N_{\alpha-\alpha} = \phi \cdot S(\alpha_\Omega)[S(\alpha_\alpha) + S(\alpha_\beta) + S(\alpha_\gamma)] / \hat{S}_\Omega$$

- Can be checked against experimental measurements
- A single value of $S(\alpha_\Omega)$ gives consistent fit to height of solute-solute nearest-neighbor peak and $N_{\alpha-\alpha}$ for $Ni_{81}B_{19}$, $Fe_{80}B_{20}$, $Ni_{80}P_{20}$, $Zr_{65}Ni_{35}$ and $Nb_{60}Ni_{40}$ but not for $Ni_{63}Nb_{37}$ or $Al_{90}Y_{10}$

Partial coordination of Ω around α ($N_{\alpha-\Omega}$)

- Given directly from N(R^*) for binary glasses when $S(\alpha_\Omega)$=0
- Non-integer values of $N_{\alpha-\Omega}$ are anticipated for $R \approx R^*$ via the concept of quasi-equivalent clusters (Sheng *et al., Nature*, **439**, 2006 419)
- When (α_Ω) defects are present,

$$N_{\alpha-\Omega} = N(R^*) - N_{\alpha-\alpha}$$

N	R^*
8	0.617
9	0.710
10	0.799
11	0.884
12	0.902
13	0.976
14	1.047
15	1.116
16	1.183
17	1.248
18	1.311
19	1.373

SHORT RANGE ORDER
Predictions vs. Experiment

Experimental and predicted partial coordination numbers

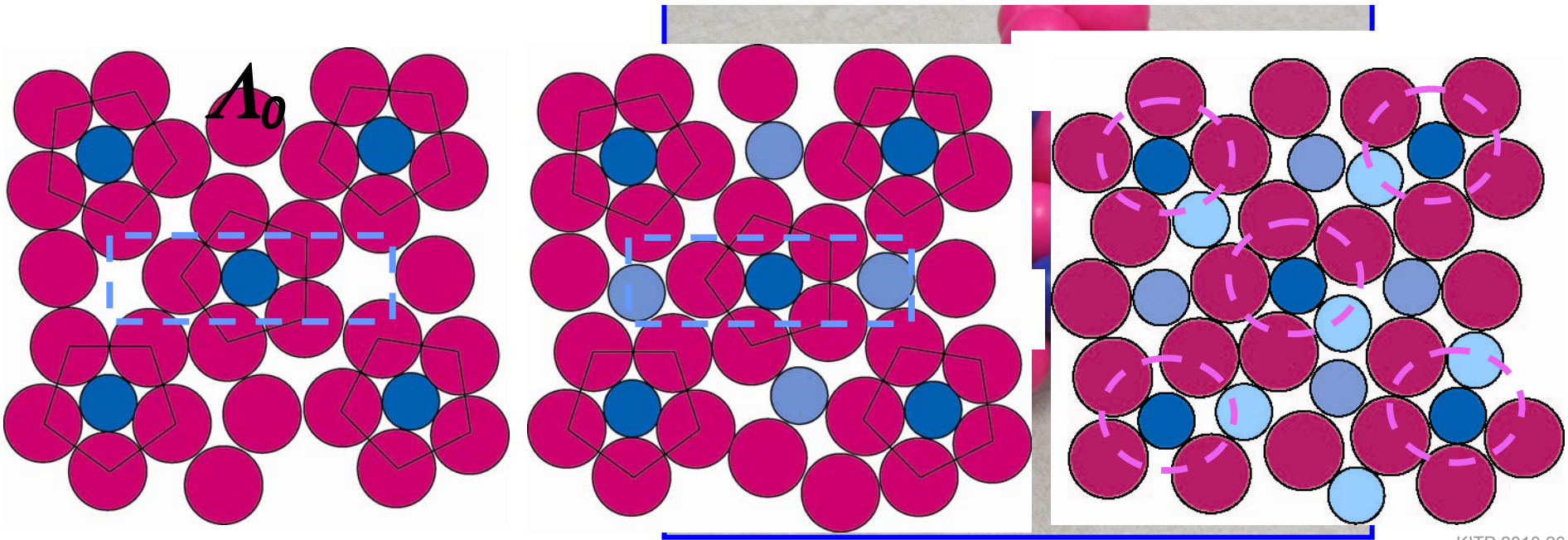
Glass		$N_{\Omega-\Omega}$	$N_{\Omega-\alpha}$	$N_{\alpha-\alpha}$	$N_{\alpha-\Omega}$	Ref.
$Ni_{81}B_{19}$ $\langle 9\rangle_{sc}$	Expt.	10.8	2.2	0	9.3	[4]
	Pred.	10.0	1.93	0	8.62	
$Ni_{64}B_{36}$ $\langle 9\rangle_{sc}$	Expt.	9.2	4.9	1.1	8.7	[84]
	Pred.	9.07	4.78	1.94	6.67	
$Fe_{80}B_{20}$ $\langle 9\rangle_{sc}$	Expt.	12.4	2.16	0	8.64	[85]
	Pred.	10.0	2.11	0	8.52	
$Ni_{80}P_{20}$ $\langle 10\rangle_{sc}$	Expt.	9.4	2.33	0	9.3	[76]
	Pred.	10.00	2.50	0	9.80	
$Zr_{65}Ni_{35}$ $\langle 10\rangle_{sc}$	Expt.	9.0	2.9	2.3	5.4	[74]
	Pred.	7.50	2.59	1.88	8.65	
$Nb_{60}Ni_{40}$ $\langle 12\rangle_{fcc}$	Expt.	9.0	5.5	3.8	8.2	[72]
	Pred.	8.63	6.21	3.44	7.03	
$Ni_{63}Nb_{37}$ $\langle 16\rangle_{fcc}$	Expt.	6.6	5.9	5.6	10.0	[72]
	Pred.	10.19	6.56	5.23	11.40	
$Al_{90}Y_{10}$ $\langle 17\rangle_{fcc}$	Expt.	10.7 ± 0.8	1.6 ± 0.2	1.2 ± 0.9	14.2 ± 1.3	[86]
	Pred.	11.81	1.47	0.13	17.04	

Miracle, *Acta mater.*, **54**, 4317 (2006)

SOLUTE-SOLUTE MRO

Cluster organization can be studied experimentally

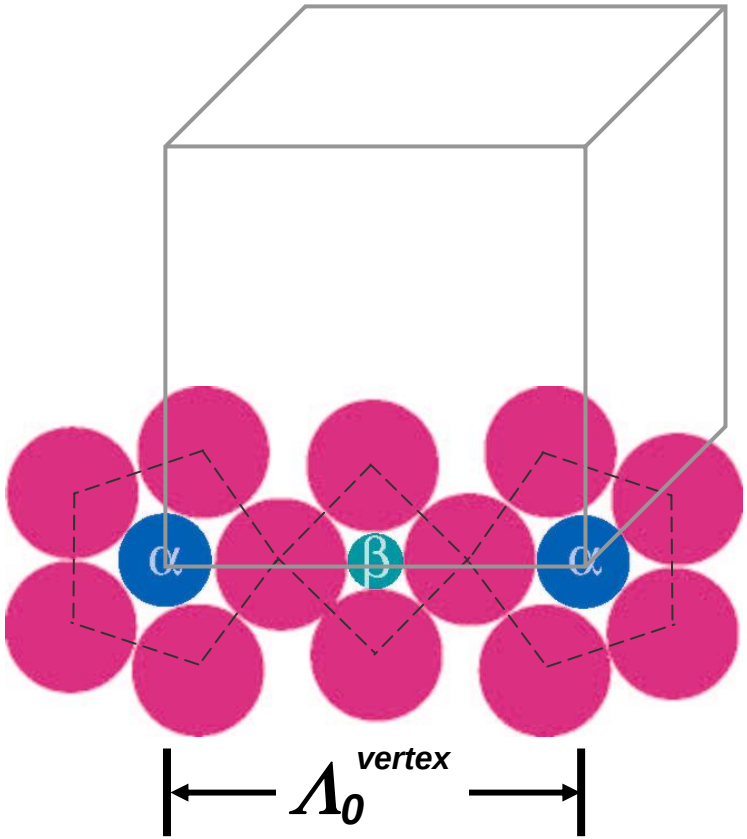
- Consider a specific cluster-packing arrangement
  - *fcc, bcc, sc, hcp, icosahedral and dense random*
- Compute the structure factor of the solute atoms alone
  - *The length scale is the cluster unit cell length, Λ_0, which can be estimated from the packing of hard spheres*
- Compare predicted structure factors with **solute-solute** partial pair distribution functions

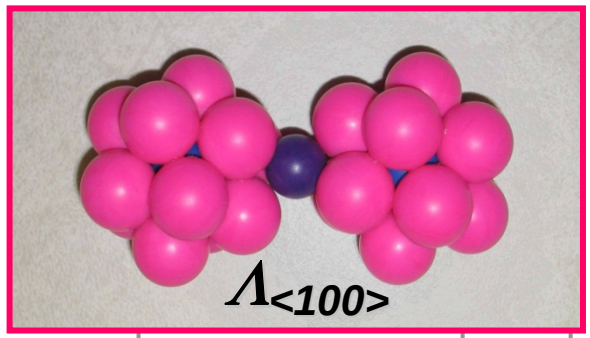


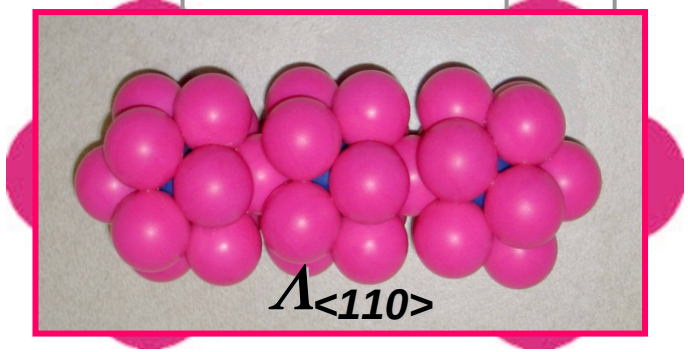
CLUSTER UNIT CELL LENGTH

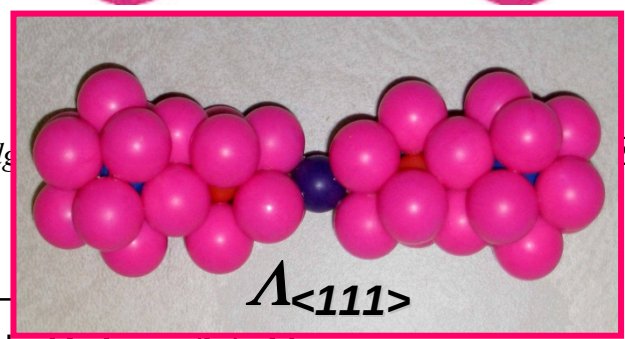
Cluster Unit Cell Length (Λ_0)

Cluster unit cell length (Λ_0) can be calculated from geometry









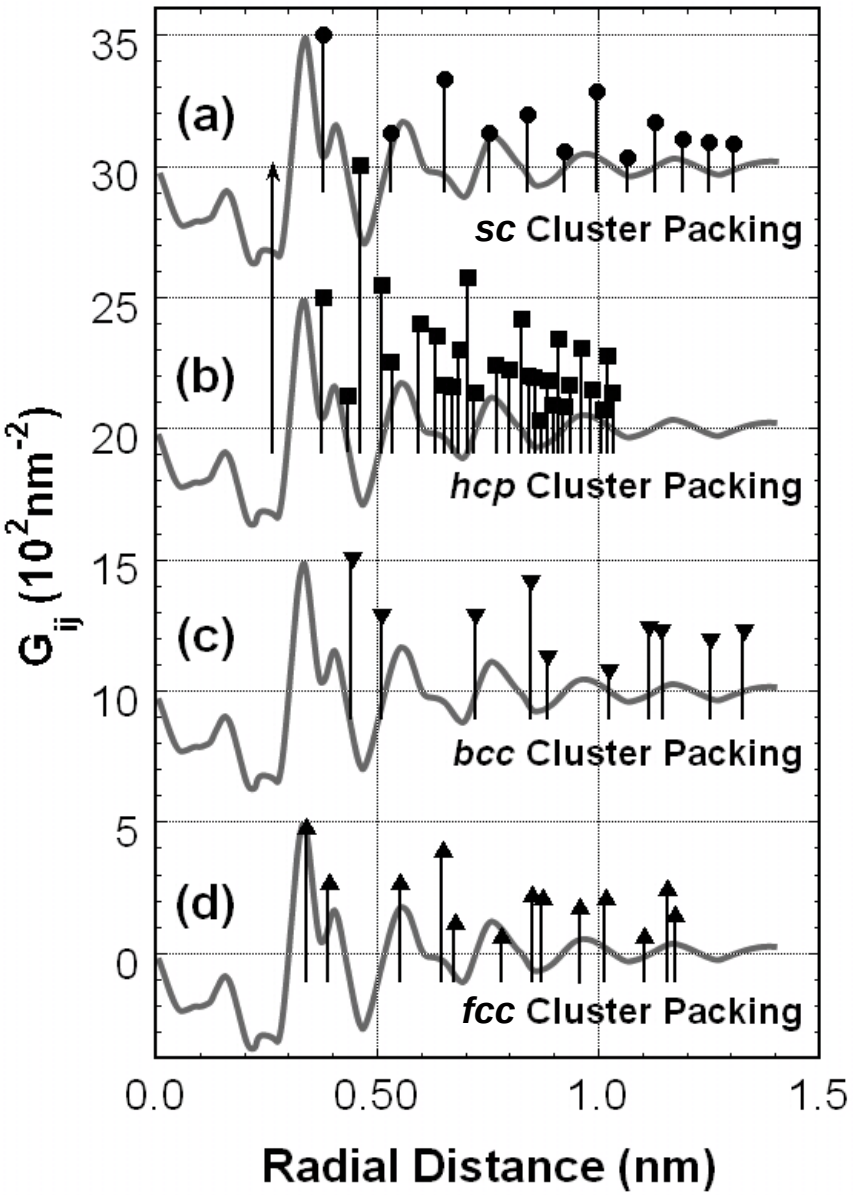
$$\Lambda_0^{vertex} = 2[R_\alpha + 2 + R_\beta]$$

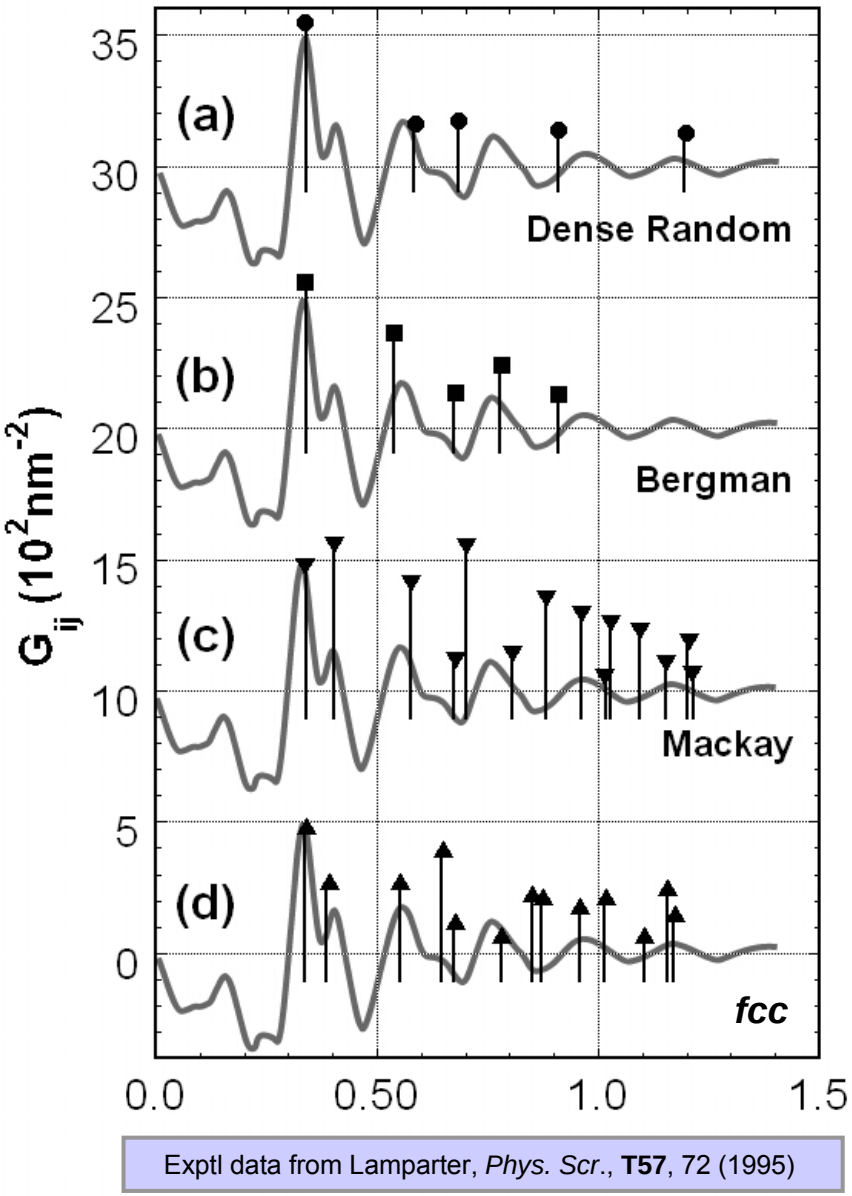
$$\Lambda_0^{edge} \quad \overline{\quad} \quad -1]$$

$$\Lambda_0^{face} = 2[\sqrt{(1+R_\alpha)^2 - 4/3} + \sqrt{(1+R_\beta)^2 - 4/3}]$$

CLUSTER PACKING SYMMETRY

B-B Partial PDF for $Ni_{81}B_{19}$



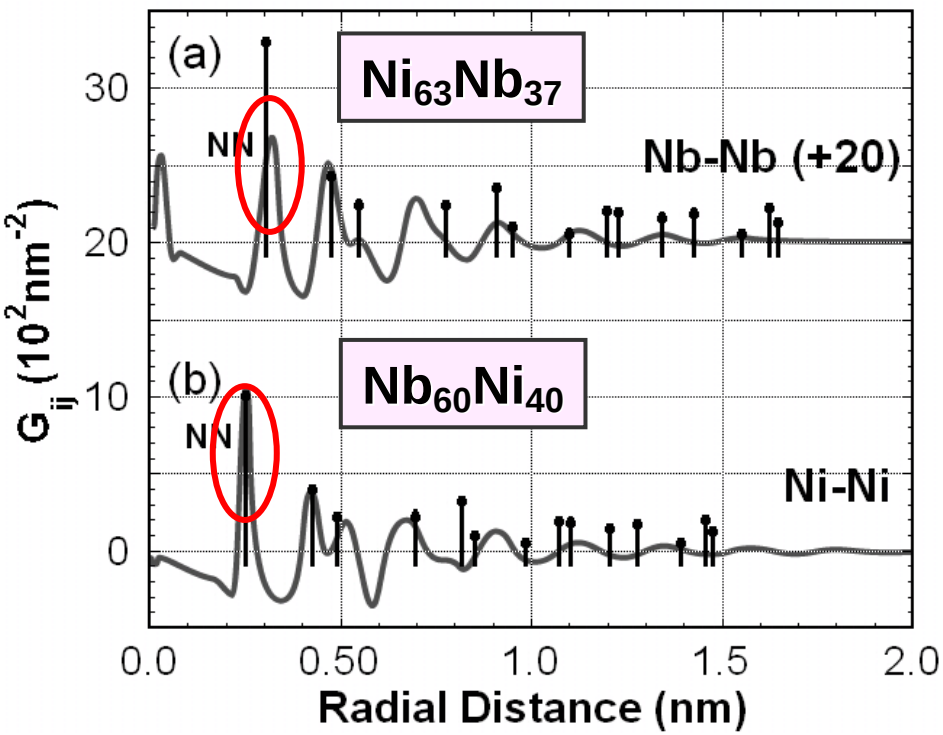


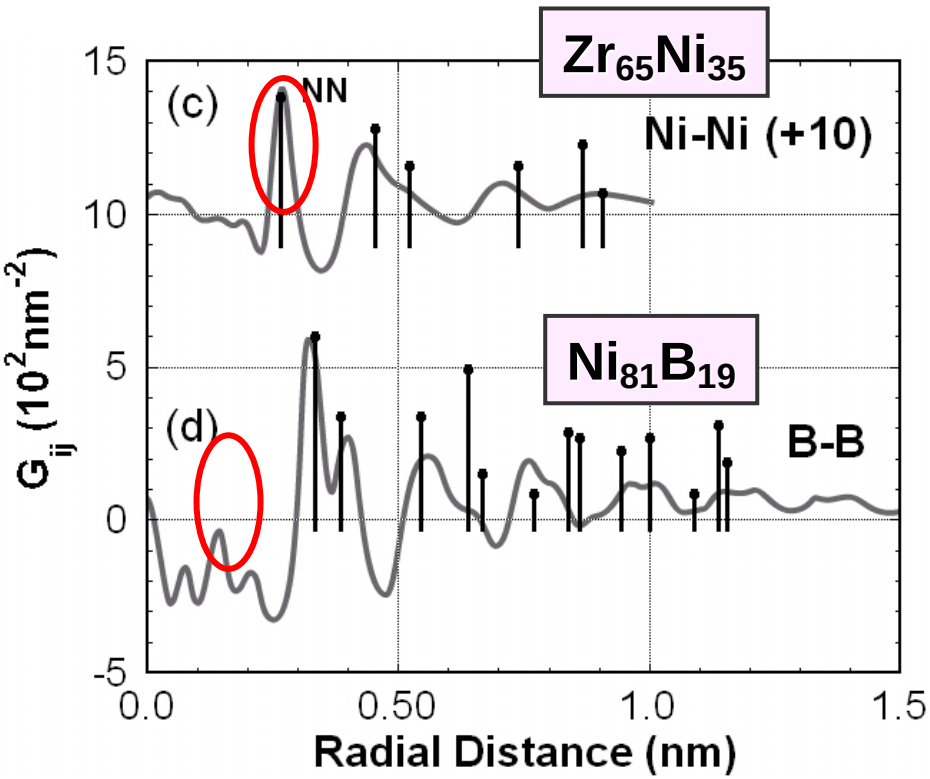
Exptl data from Lamparter, *Phys. Scr.*, **T57**, 72 (1995)

MEDIUM RANGE ORDER
Prediction

Notable results

- *fcc* cluster packing with (α_β+α_γ) defect state for $N \geq 12$, *sc* cluster packing for $N \leq 10$
- MRO for $Ni_{81}B_{19}$ now well-predicted by ECP model
- Good fit to MRO of solutes to radial distances of ~1nm





Miracle, *Nature Mat.*, **3**, 697 (2004)

DENSITY

Calculated densities and packing fractions

Composition	Actual density (g/cm^3)	Predicted density (g/cm^3)	Density error (%)	Λ_0 Error (%)	Corrected Λ_0 (nm)	Packing fraction	Ref.
$Ni_{81}B_{19}$	8.4	10.92	30.0	−9.1	0.395	0.7420	[4]
$Pd_{79.8}Si_{20.2}$	10.25	13.45	31.2	−9.5	0.663	0.6997	[52]
$Al_{85}Gd_6(Fe_3Ni_6)$	3.51	3.07	−12.6	4.4	0.928	0.7048	[19]
$Al_{85}Gd_8(Fe_2Ni_5)$	3.71	3.23	−12.9	4.5	0.927	0.7223	[19]
$Al_{87}Gd_6(Fe_1Ni_6)$	3.47	2.95	−14.9	5.3	0.920	0.7115	[19]
$Fe_{70}Zr_{10}B_{20}$	7.23	9.52	31.6	9.6	0.855	0.7086	[87]
$Fe_{70}Nb_{10}B_{20}$	7.68	9.07	18.2	−5.7	0.807	0.7183	[87]
$Fe_{70}Cr_{10}B_{20}$	7.34	7.75	5.6	−1.8	0.755	0.7105	[87]
$Mg_{60}Y_{10}Cu_{30}$	3.13	2.71	−13.4	4.7	1.039	0.6784	[88]
$Pd_{77.5}Cu_6Si_{16.5}$	10.48	9.74	−7.1	2.4	0.766	0.7051	[52]
$Pd_{48}Ni_{32}P_{20}$	9.83	6.94	−29.4	11.0	0.768	0.7227	[52]
$Pt_{52.5}Ni_{22.5}P_{25}$	15.85	13.29	−16.1	5.7	0.809	0.6811	[52]
$Zr_{60}Al_{10}Cu_{30}$	6.72	4.60	−31.6	11.9	0.912	0.7254	[21]
$Zr_{60}Al_{15}Ni_{25}$	6.36	4.07	−36.0	13.8	0.896	0.7272	[21]

Miracle, *Acta mater.*, **54**, 4317 (2006)

OUTLINE

LOCAL STRUCTURE

EXTENDED STRUCTURES

BINARY METALLIC GLASS STRUCTURE

BINARY METALLIC GLASS ASSESSMENT

Compare Topology vs Stability/GFA

SUMMARY

Miracle, Louzguine, Louzguina, Inoue; *Inter. Mater. Review*, In press.

TOPOLOGICAL ASSESSMENT

Approach

Extensive literature review of binary metallic glasses

- Tabulate constitution of known binary glasses
  - *Include only glasses produced by quenching from the liquid state*
  - *Nearly 200 citations, giving 628 distinct alloys in 175 binary systems*
- Document T_g, T_x *and* T_{liq} and derived parameters T_{rg}, $\Delta T_x = T_x - T_g$, T_x / T_{liq} and $\gamma_{th} = T_x / (T_g + T_{liq})$
- Document maximum reported thickness
- Document structure-specific information such as density and partial coordination numbers

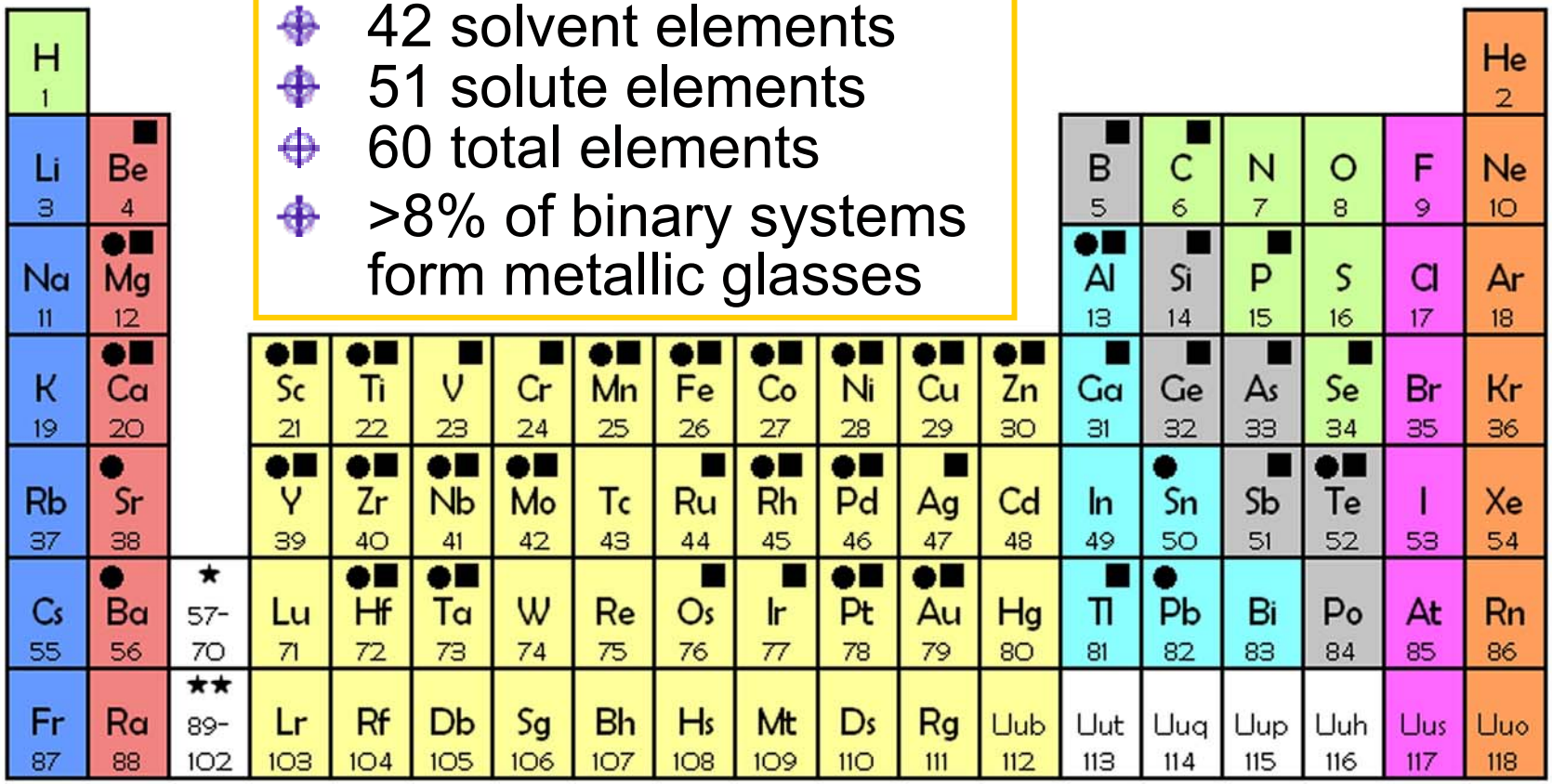
Characterize topology with the ECP model

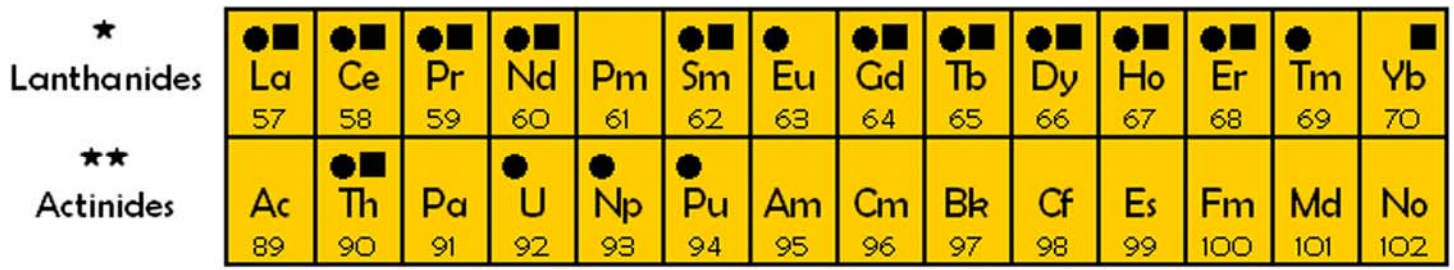
- The relative number and sizes of constituent atoms
- The location of atoms in the structure
- The type and concentrations of defects
- Other parameters of suggested importance
  - *Elastic properties, electronegativity…*

Assess measured stability against structural characteristics

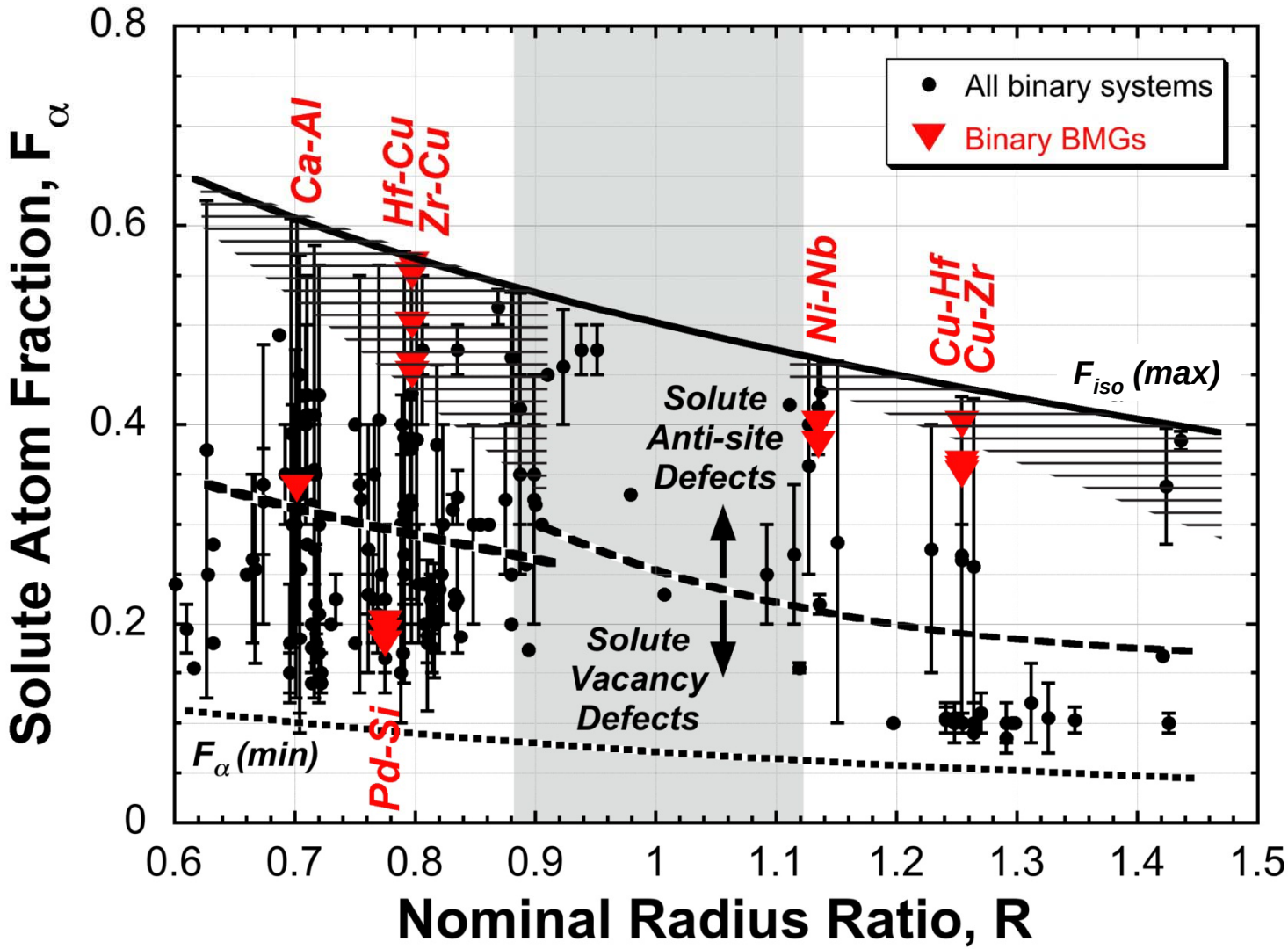
ELEMENTS FOUND IN BINARY METALLIC GLASSES

- 42 solvent elements
- 51 solute elements
- 60 total elements
- >8% of binary systems form metallic glasses





BINARY GLASS TOPOLOGIES



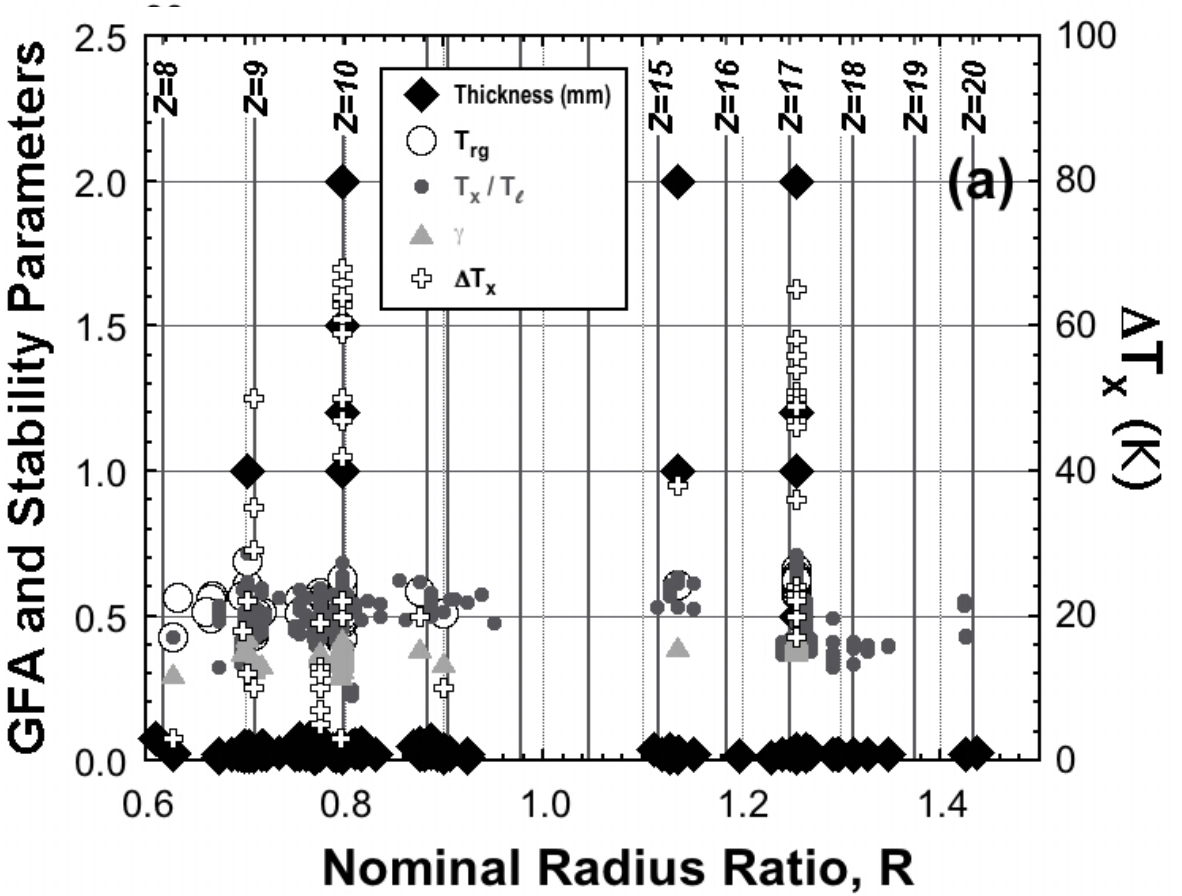
Miracle, Louzguine, Louzguina, Inoue; *Inter. Mater. Review*, In press.

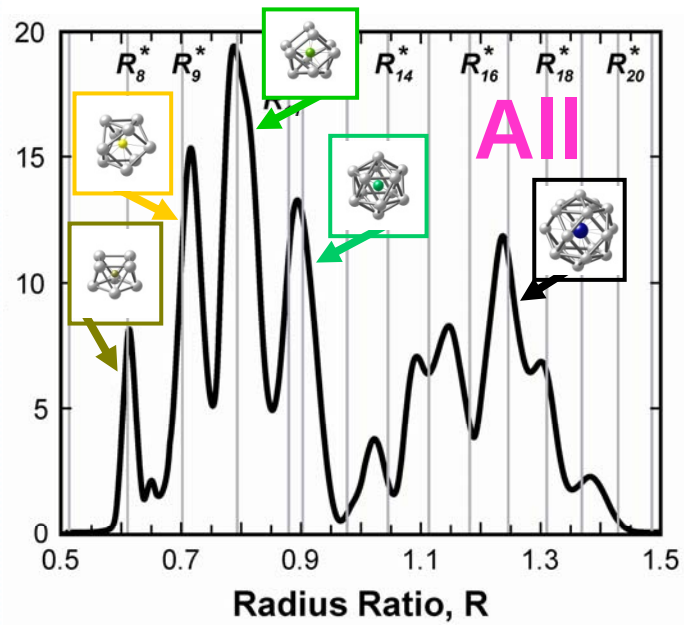
RADIUS RATIOS

Of 15 possible R values, only 5 are common

– R = 0.710, 0.799, 0.902, 1.116, 1.248

– Other values of R are uncommon or are not observed





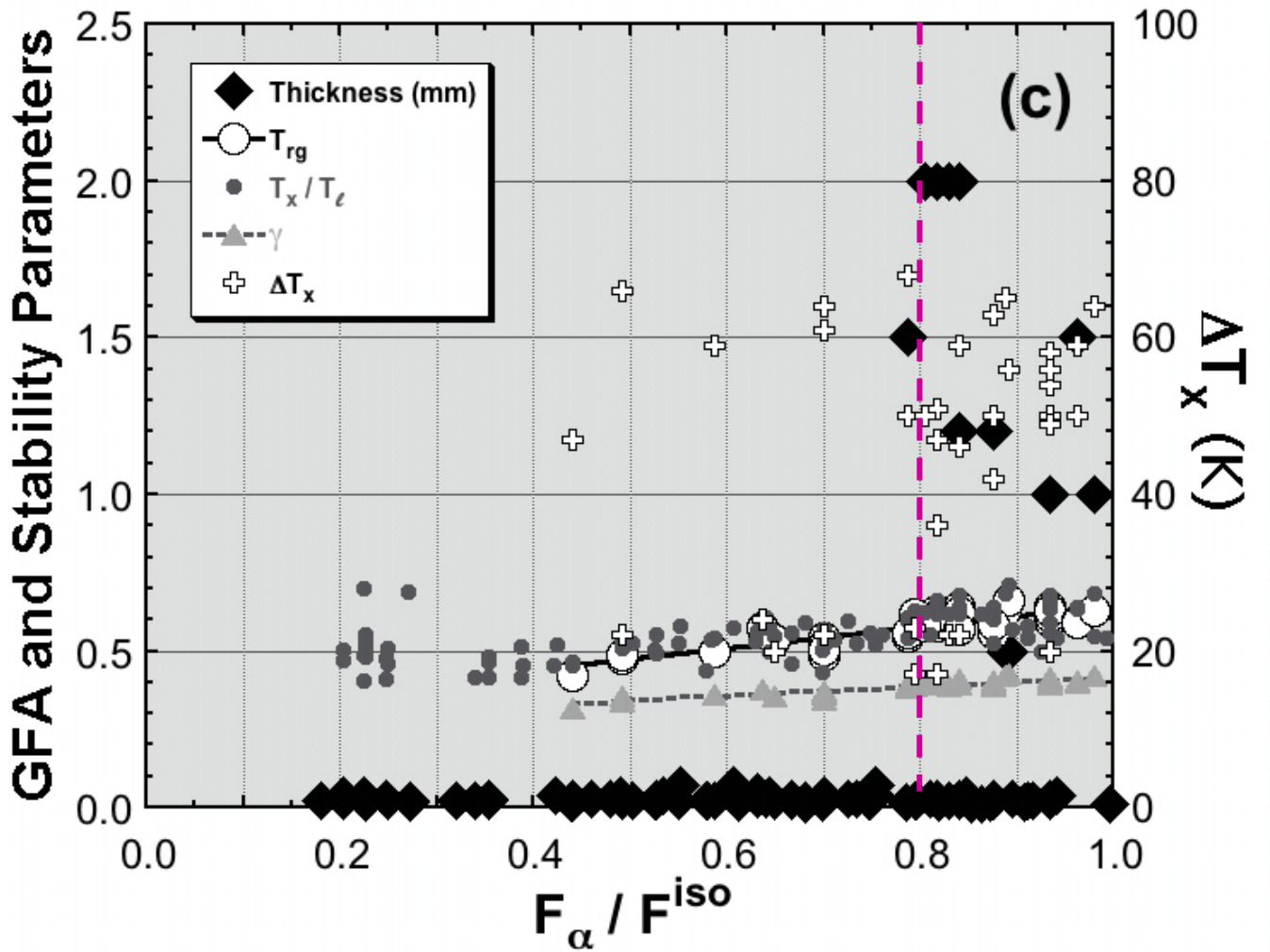
Miracle, Sanders and Senkov; *Phil. Mag.*, **83A**, 2409 (2003)

STABILITY v STRUCTURE

Binary BMGs Are Solute-Rich

All glasses but one have $\overline{S}_\alpha > 1$: All α sites are filled by α

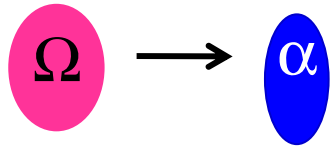
All BMGs (except Pd-Si) have $\overline{S}_\alpha > 4$: α_Ω defects are common



SOLUTE-RICH STABILITY
Contribution of Bond Enthalpy, ε

Replace a solvent atom with a solute atom

- Breaks 1 α–Ω bond and 5 Ω–Ω bonds
- Makes 1 α–α bond and 5 α–Ω bonds



A net energy change is produced by this α_Ω antisite defect

$$\Delta H = (\mathcal{E}_{\alpha\alpha} + 5\mathcal{E}_{\alpha\Omega}) - (\mathcal{E}_{\alpha\Omega} + 5\mathcal{E}_{\Omega\Omega}) = \mathcal{E}_{\alpha\alpha} + 4\mathcal{E}_{\alpha\Omega} - 5\mathcal{E}_{\Omega\Omega}$$

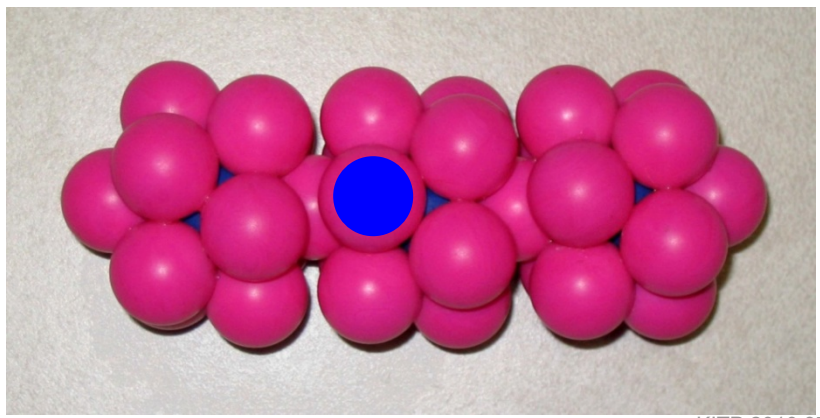
- Different solute sizes give different changes in bond numbers

Antisite defects are favored when $\Delta H < 0$

- This can occur if $\mathcal{E}_{\alpha\Omega}$ is sufficiently more negative than $\mathcal{E}_{\alpha\alpha}$ and $\mathcal{E}_{\Omega\Omega}$

$$4\mathcal{E}_{\alpha\Omega} < 5\mathcal{E}_{\Omega\Omega} - \mathcal{E}_{\alpha\alpha}$$

- This can give a stabilizing contribution in selected solute-rich glasses
- Entropy increases the magnitude of this effect
- Valid for filling up to ~1/3 of the Ω sites by α_Ω defects



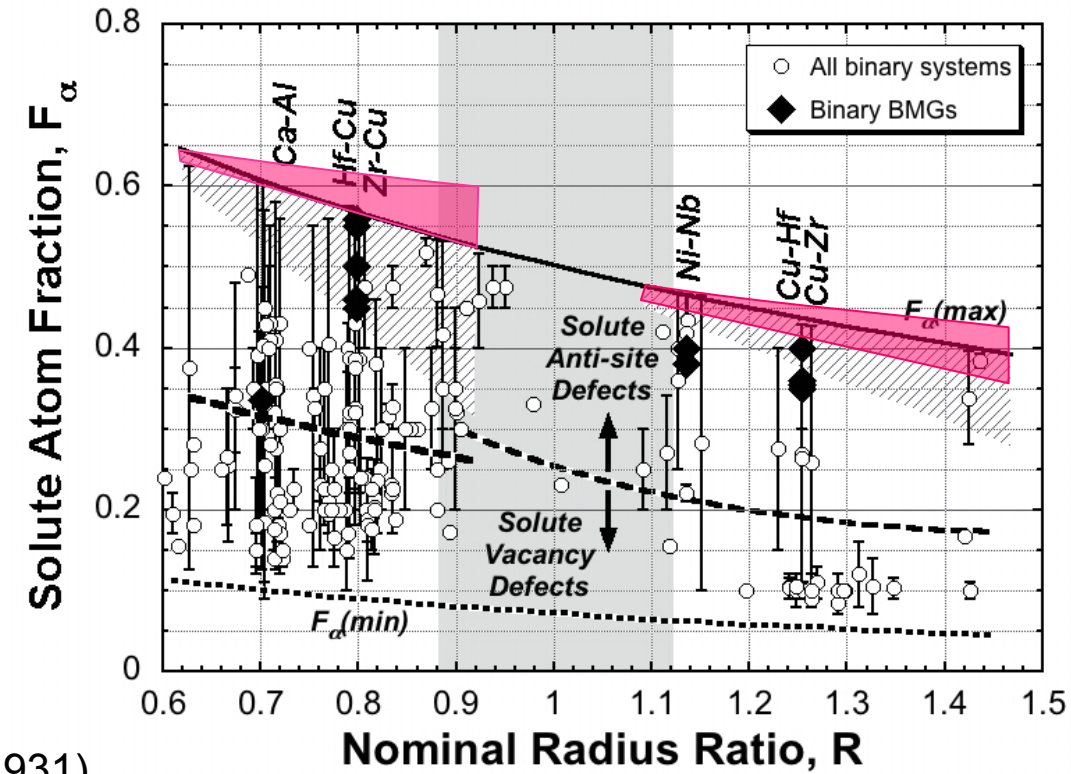
SOLUTE-RICH STABILITY
Global Packing Efficiency

The Furnas model[#] was used to estimate the atom fraction and relative size at which maximum packing occurs

- Based on filling of sphere interstices by much smaller spheres
- Extended previously to spheres with $0.1 \leq R \leq 0.5$
- Extended here to spheres with $0.6 \leq R \leq 0.9$

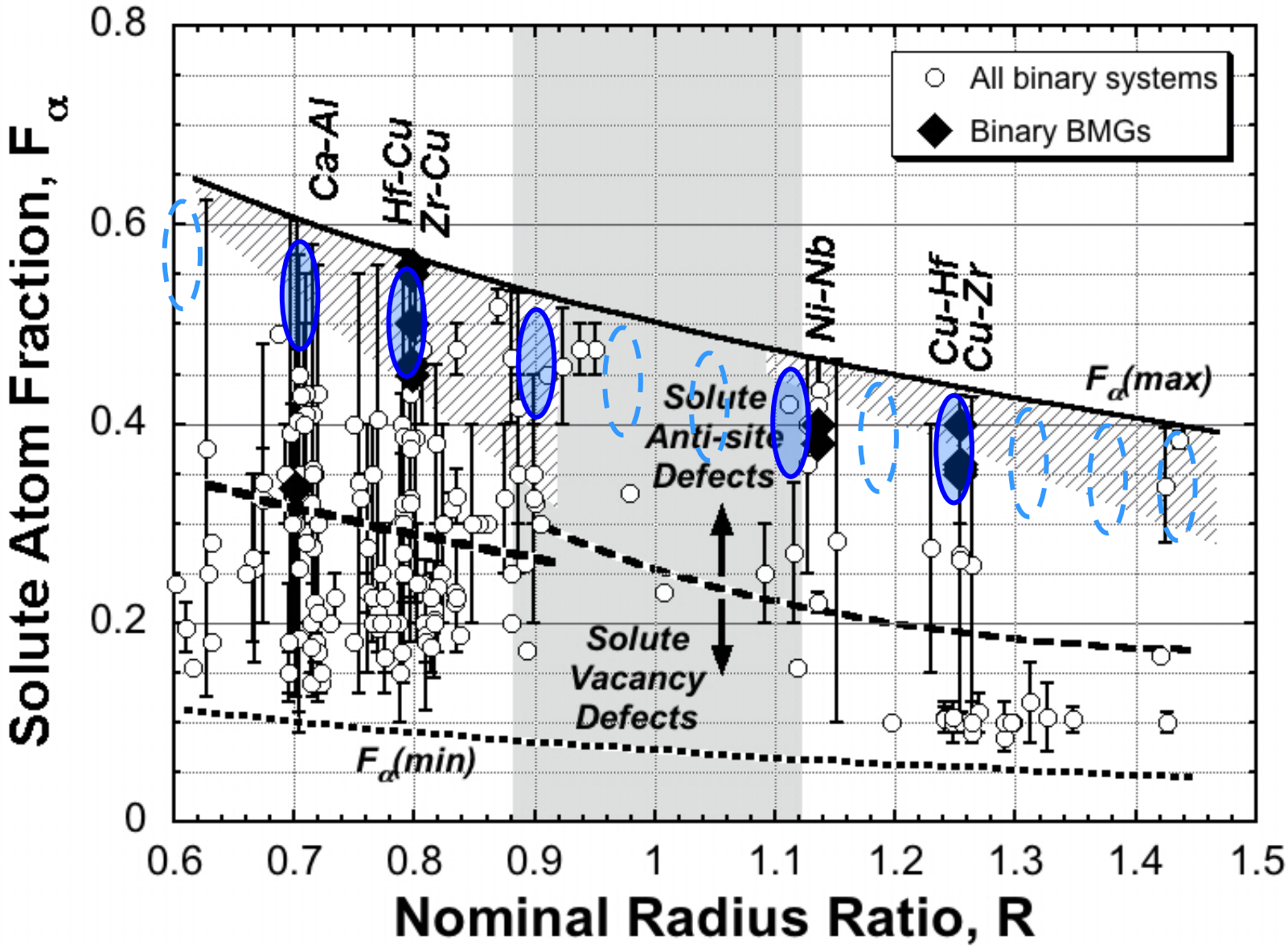
Maximal global packing efficiency occurs for solute-rich glasses near F_{iso}

- Occurs over the full range of R



[#] Furnas, *Industrial & Engr. Chem.*, **23**, (1931)

SOLUTE FRACTION v SIZE



SUMMARY

Metallic glass structures can be modeled as efficiently-packed clusters that are organized over a finite length scale

- The number of structural sites can be quantified as a function of *R*
- Structural site occupancies can be established in binary glasses
- Structural defects exist and are important
- Helps organize analysis of computer simulations

Strong preference for *R gives efficient local atomic packing**

- Only five *R* values are common (0.71, 0.80, 0.90, 1.12 and 1.25)
- 4 complementary inverse systems give the majority of glasses and BMGs
- The scarcity of many *R* values is not understood

Solute-rich compositions stabilize binary metallic glasses

- Enable efficient global atomic packing (Furnas model)
- Give α_Ω anti-site defects that maximize the number of unlike atom bonds
- Packing and chemical stability are both maximized for $F_\alpha > 0.8\ F_{iso}$

Relative atom size, solute concentration and chemistry all make important contributions to stability

- These contributions must all be satisfied simultaneously

THANK YOU!