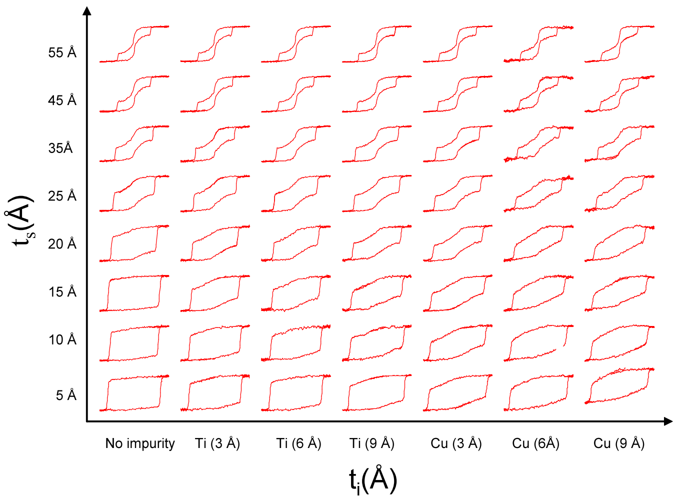
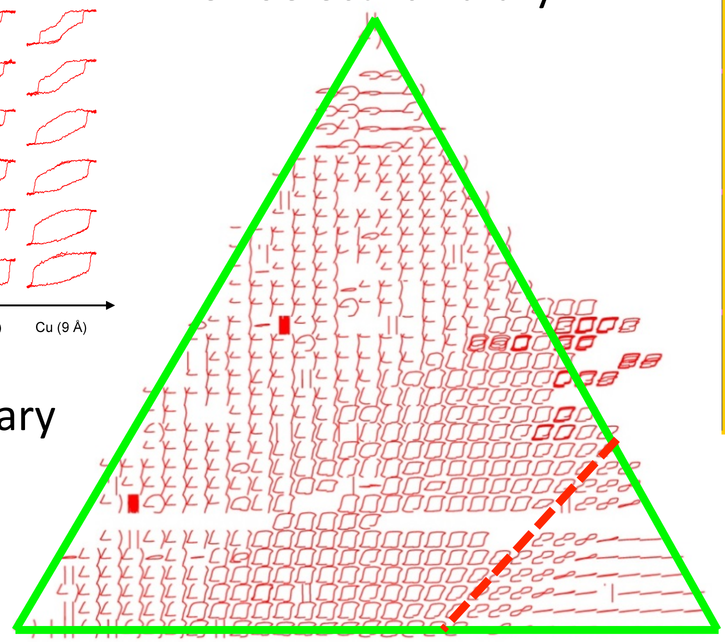
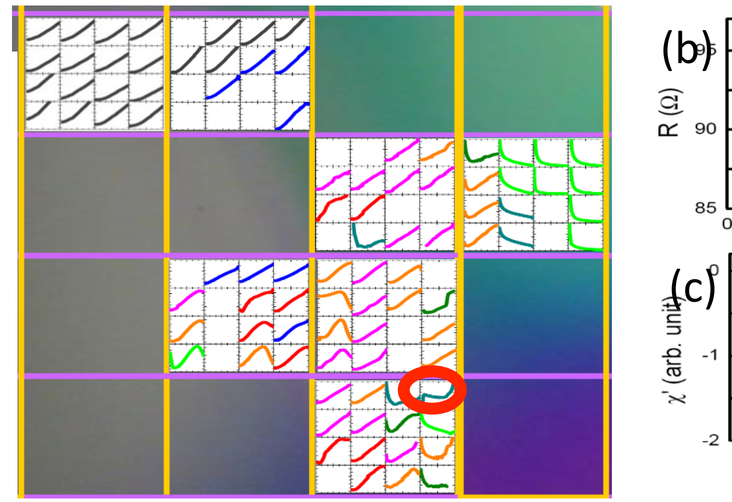
Informatics for
Combinatorial Materials Science



Permanent magnet library

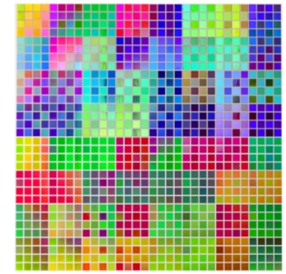
Ferroelectric library





Superconductor library

Ichiro Takeuchi
University of Maryland



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Duke
S. Curtarolo

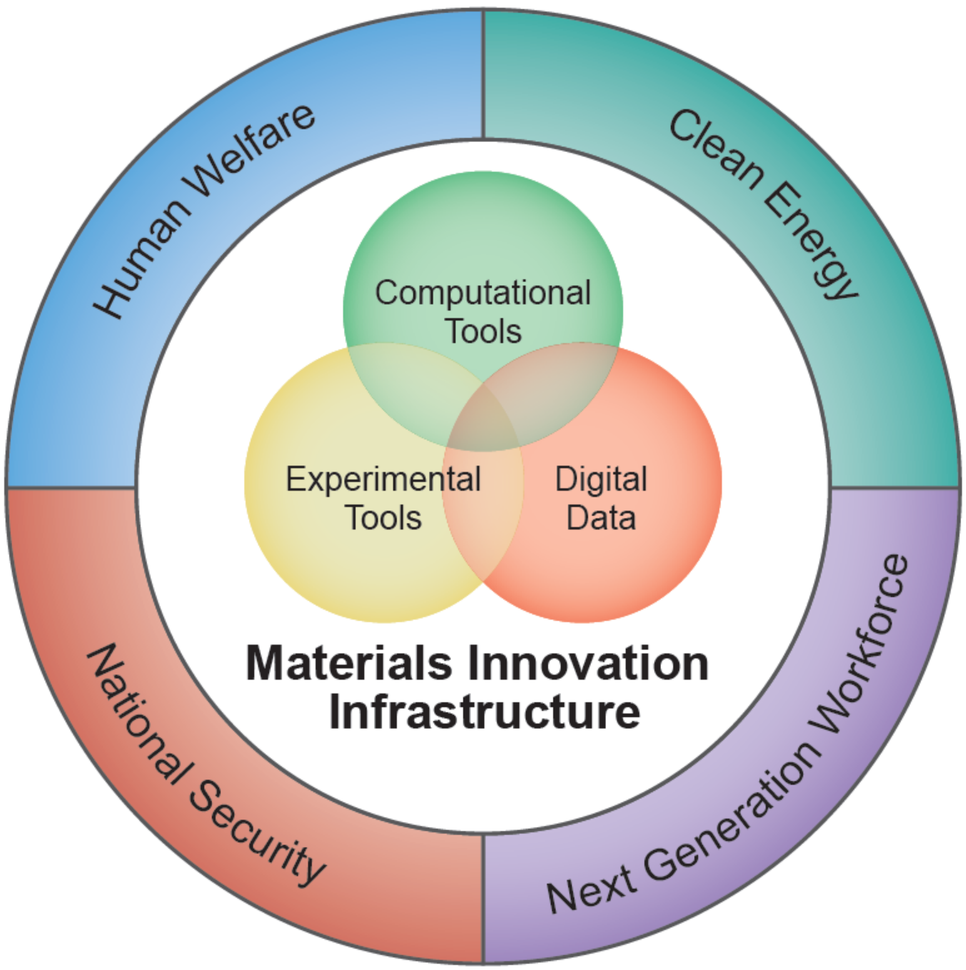
Ames Lab
M. J. Kramer

SLAC
A. Mehta

Support

DOE EERE, ARPA-E, NIST, ONR

The Materials Genome Initiative



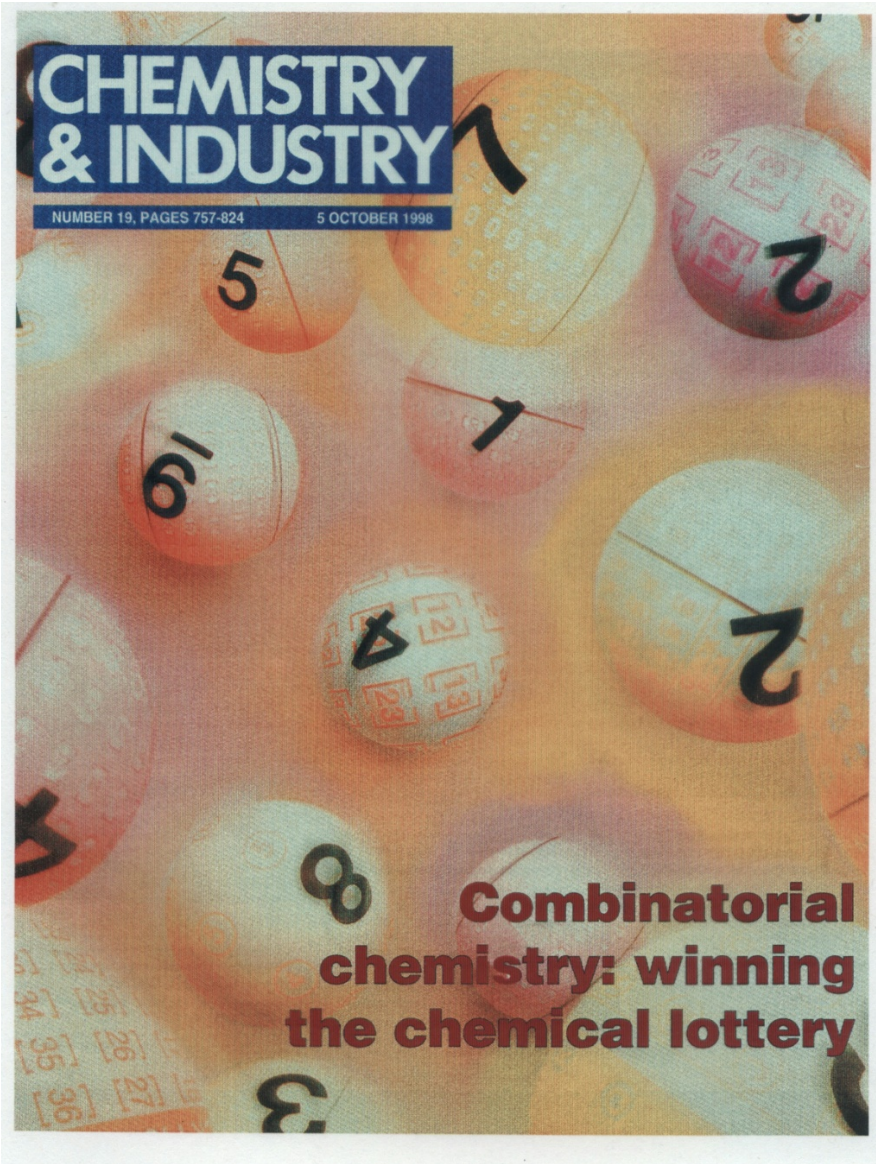
- Effective coupling between theory and experiments are needed
- Combinatorial experiments are the natural counterpart to computational efforts
- Lack of concerted effort in incorporating experiments; gap between theory and experiment

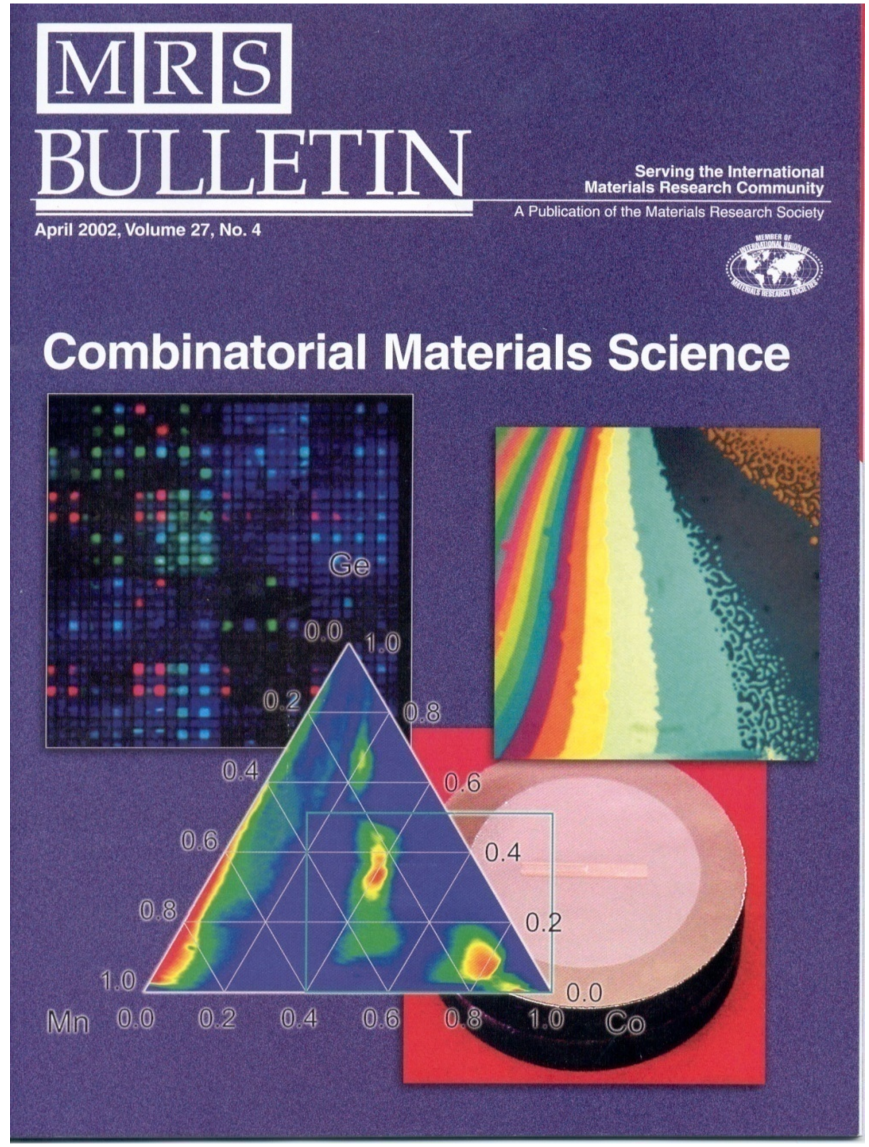
http://www.mrs.org/mgi-workshop-2014/

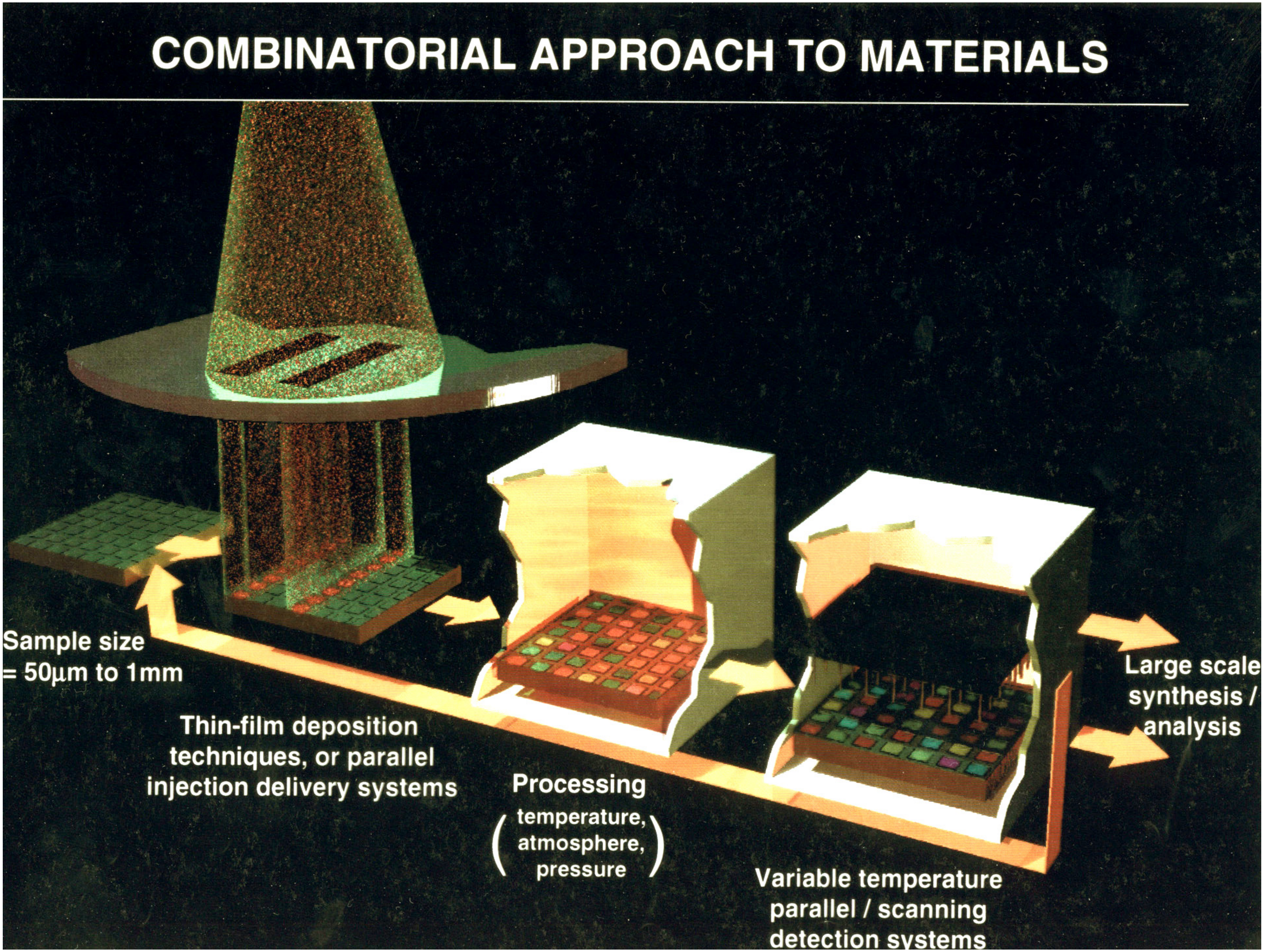
MANY AT A TIME Program head Andreas Marzinik (front to back) and lab specialists Raphael Gattlen and Urs Rindisbacher of Novartis Pharma AG, Basel, Switzerland, pipette coupling reagent into 96-well reaction blocks.

COMBINATORIAL CHEMISTRY

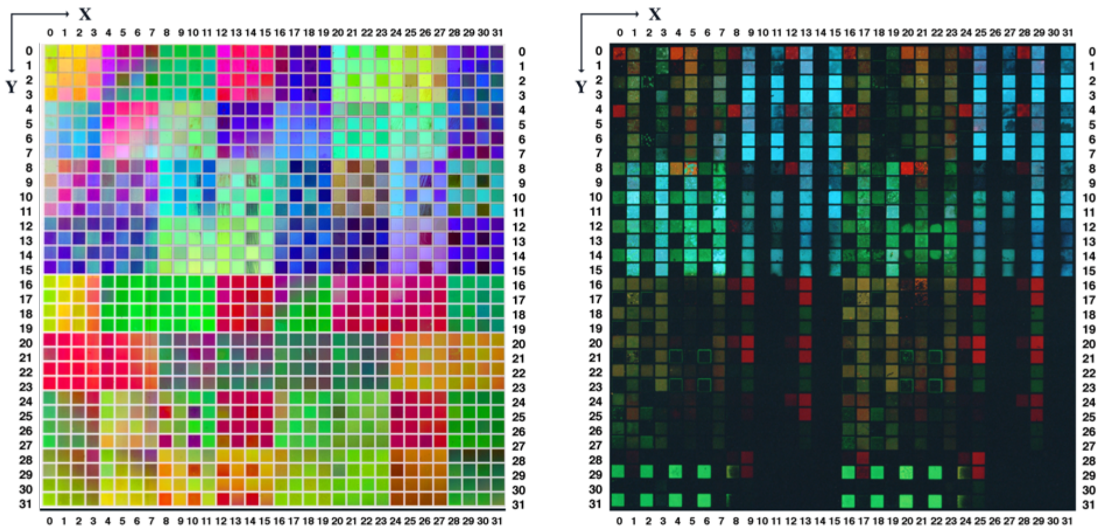
Chemical & Engineering News, August 2001


CHEMISTRY & INDUSTRY
NUMBER 19, PAGES 757-824
5 OCTOBER 1998
Combinatorial chemistry: winning the chemical lottery

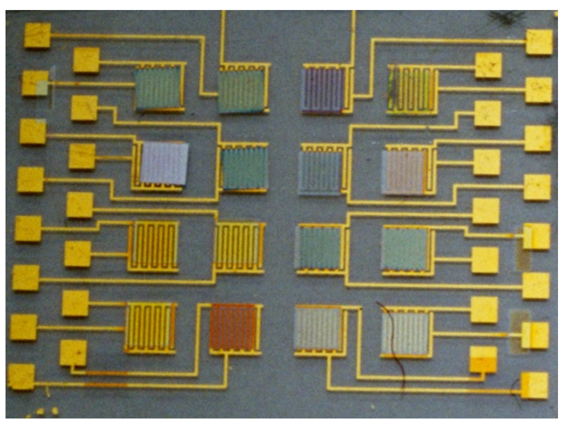

MRS BULLETIN
Serving the International Materials Research Community
A Publication of the Materials Research Society
April 2002, Volume 27, No. 4
Combinatorial Materials Science
Ge
Mn
Co


COMBINATORIAL APPROACH TO MATERIALS
Sample size = 50µm to 1mm
Thin-film deposition techniques, or parallel injection delivery systems
Processing
(temperature, atmosphere, pressure)
Variable temperature parallel / scanning detection systems
Large scale synthesis / analysis

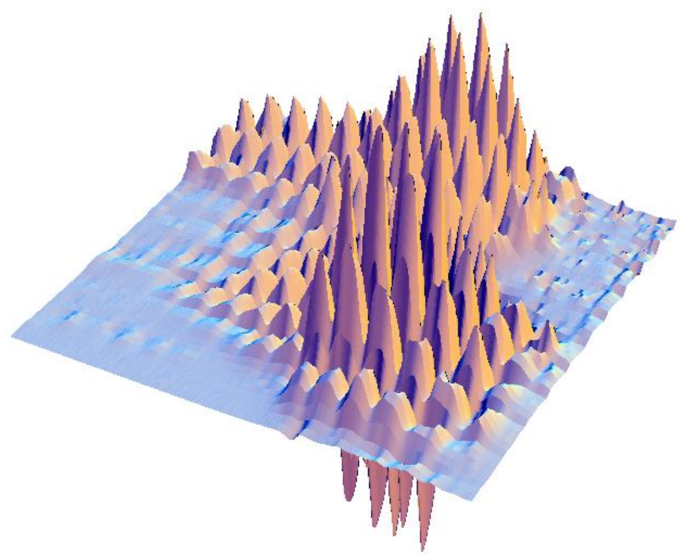
Combinatorial Libraries of Inorganic Materials



Luminescent materials libraries, Science **279**, 1712 (1998)

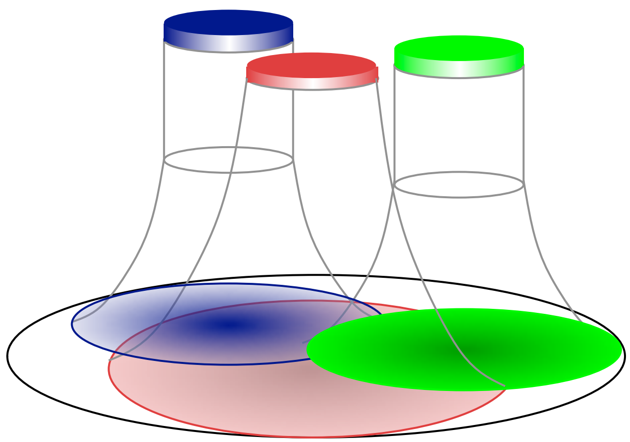


Semiconductor gas sensor library, “electronic nose”, Appl. Phys. Lett. **83**, 1255 (2003)

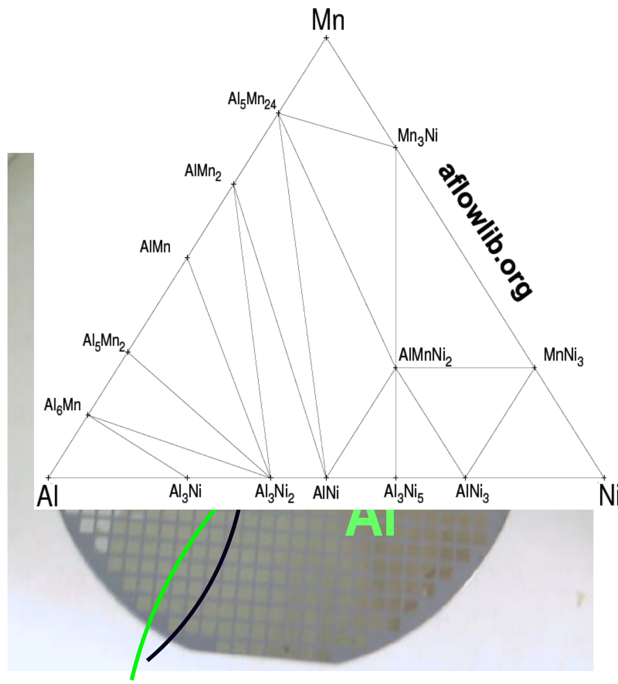


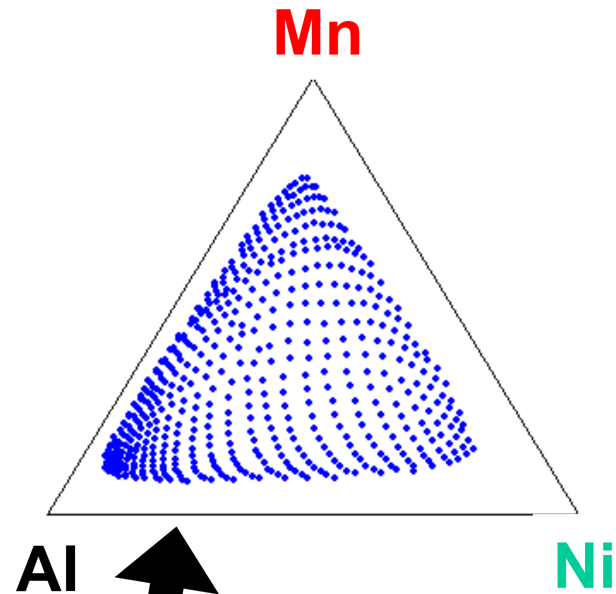
Magnetic shape memory alloy library, Nature Materials **2**, 180 (2003)

Composition Spreads of Ternary Metallic Alloy Systems



Co-sputtering scheme



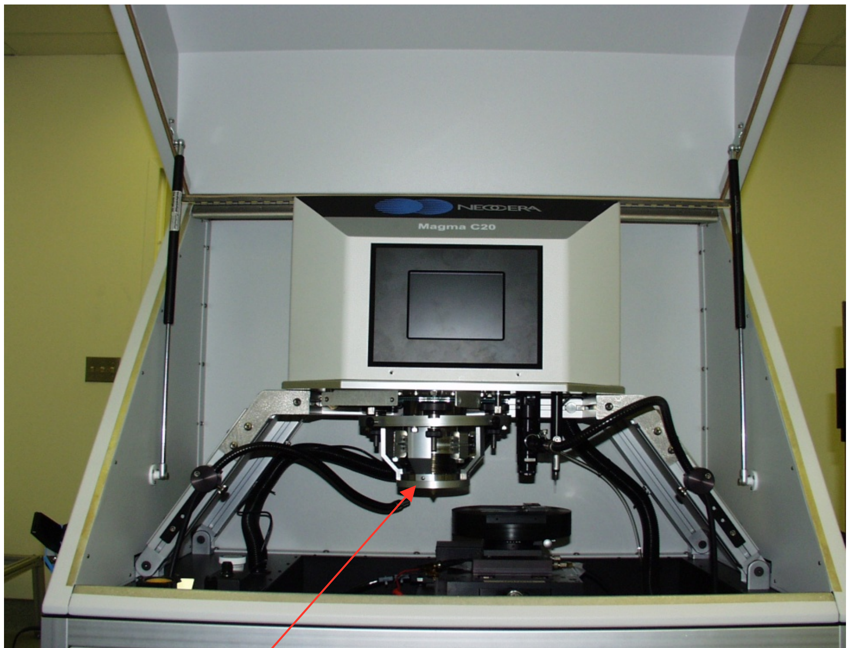


Phase diagram

Composition is mapped using an electron probe (WDS)

Review article: Green *et a*l., JAP **113**, 231101 (2013)

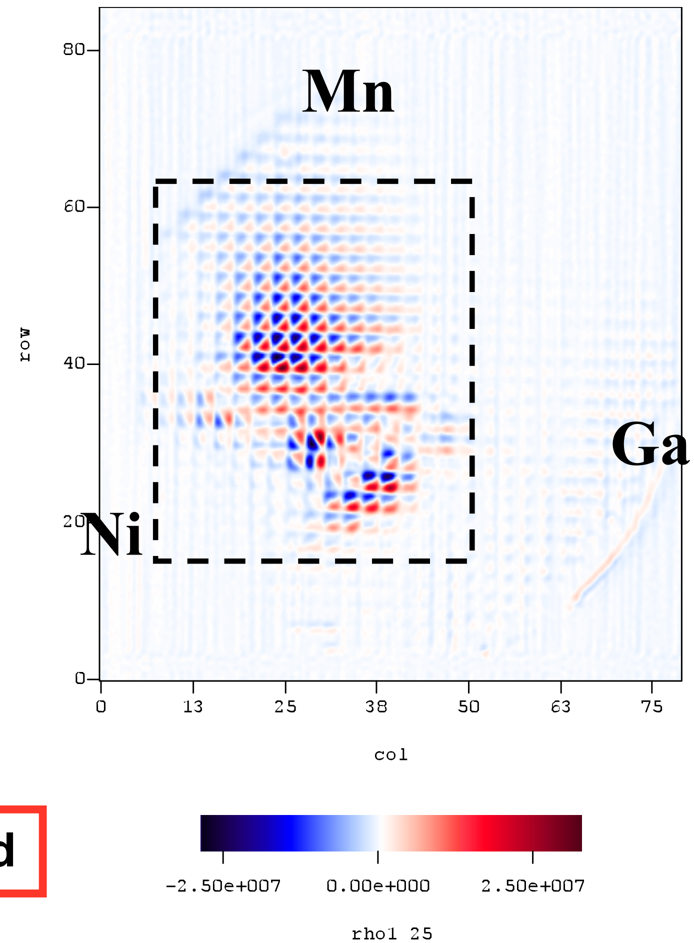
Rapid mapping of magnetic properties: scanning SQUID



SQUID assembly inside vacuum

Room temperature samples are measured

Raw data



Nature Materials **2**, 180 (2003)

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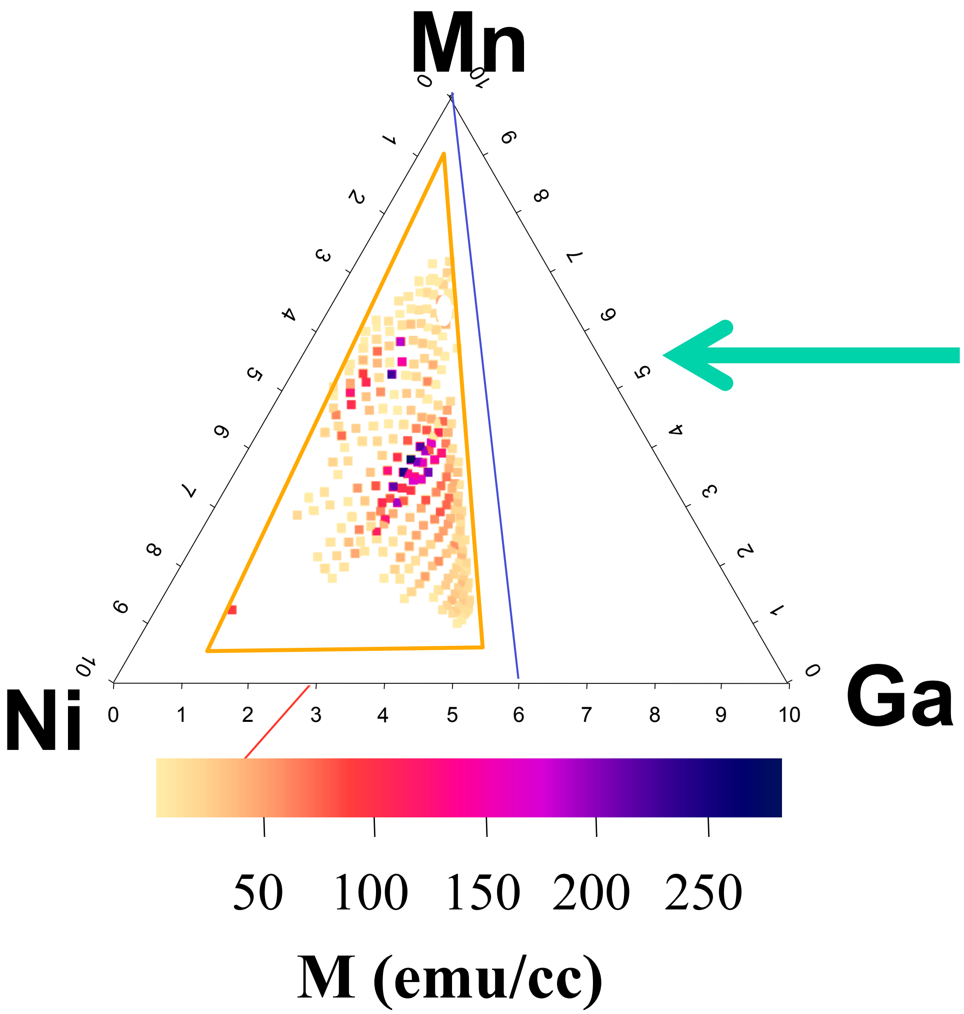
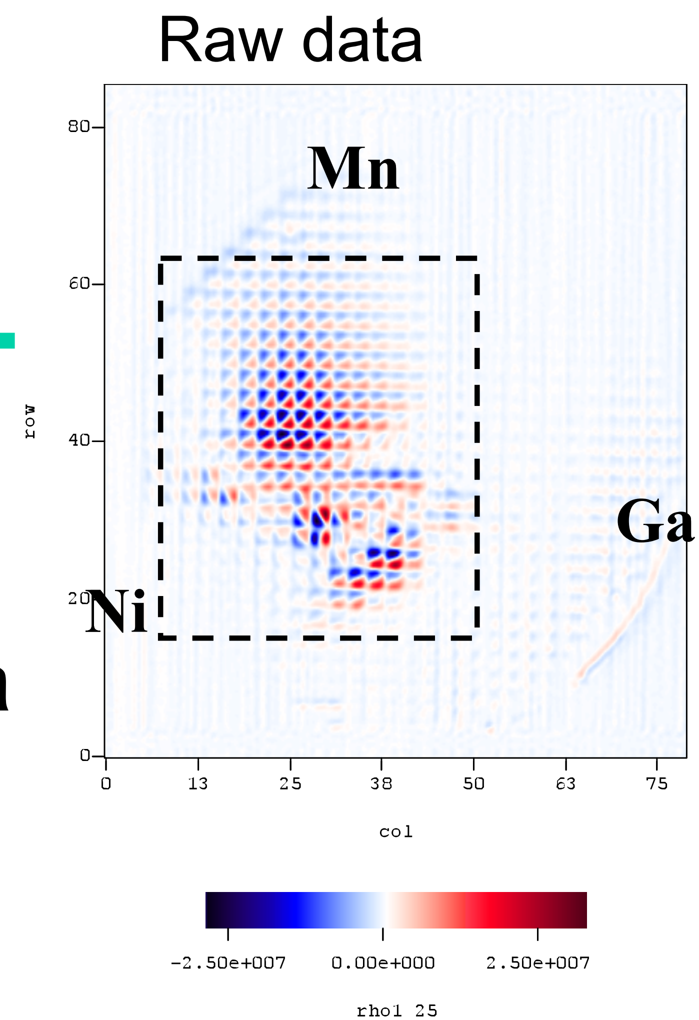
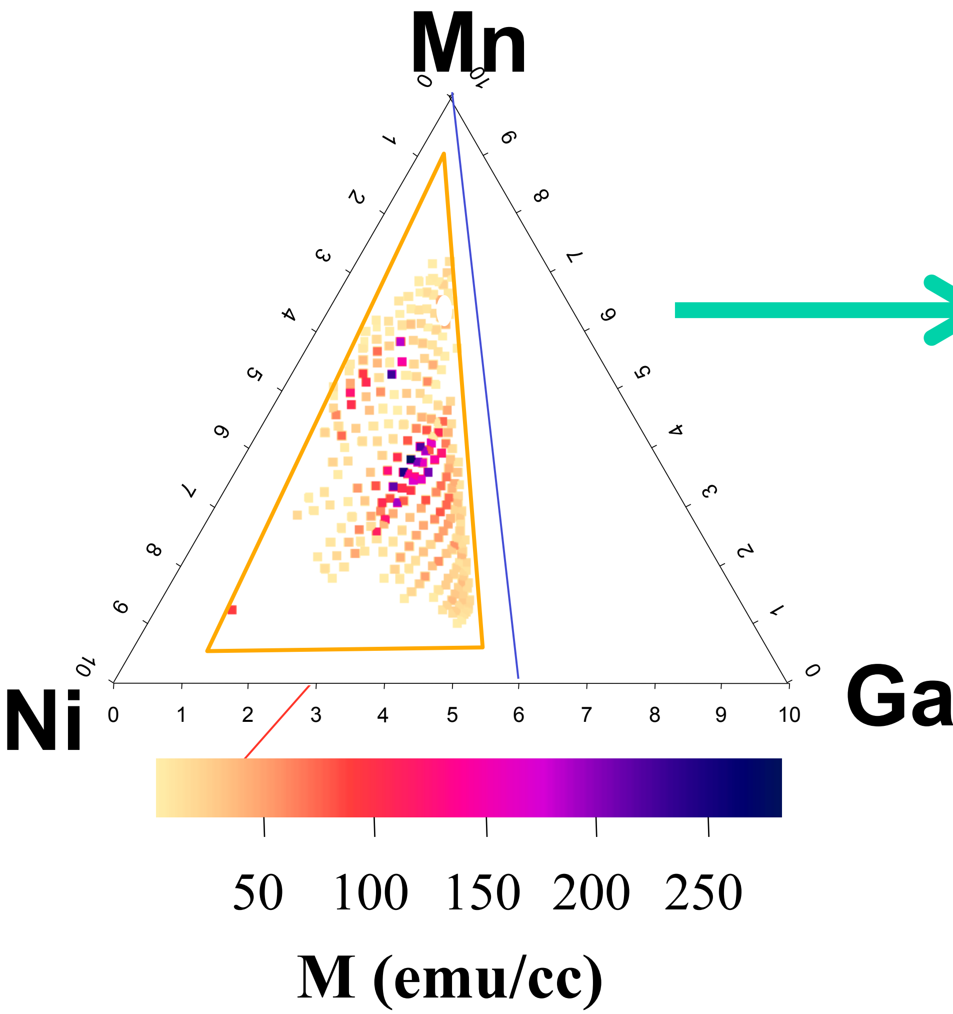
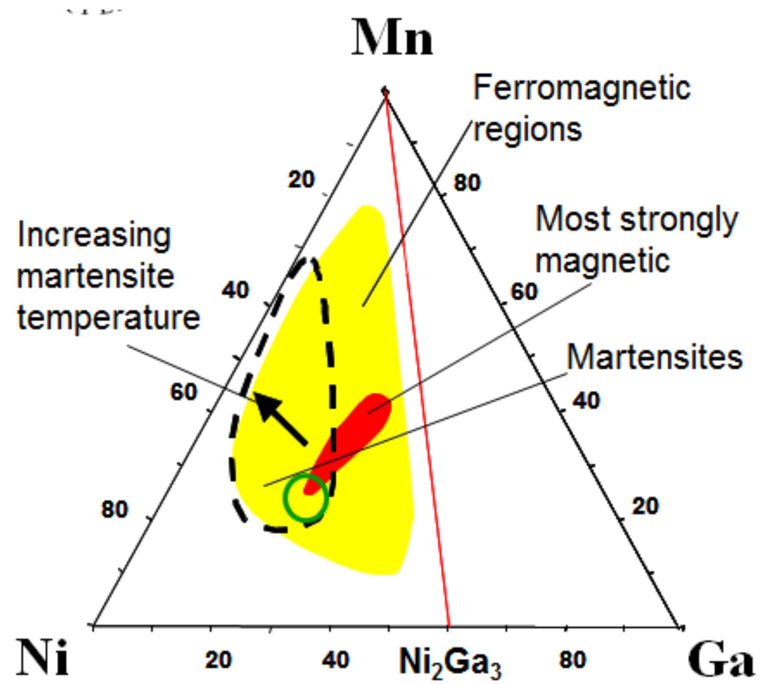




Nature Materials **2**, 180 (2003)

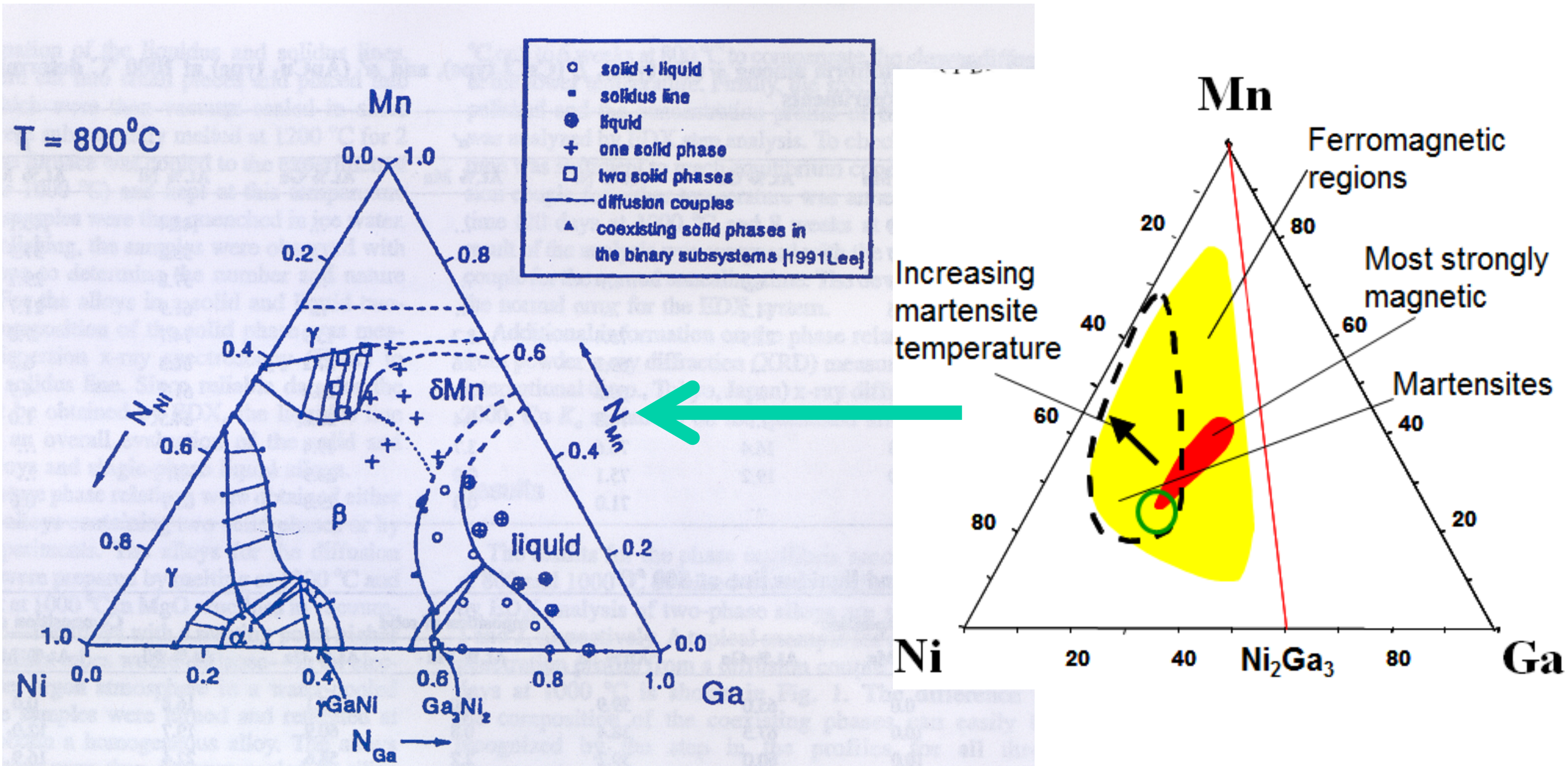
Rapid mapping of magnetic properties: constructing functional phase diagram





Nature Materials **2**, 180 (2003)

Rapid mapping of magnetic properties: comparison with phase diagram



C. Wedel and K. Itagaki,
Journal of Phase Equilibria 22, 324 (2001)

Nature Materials **2**, 180 (2003)

Data driven approaches to the combinatorial strategy

Validation of theoretical predictions (high-throughput computations)

Visualization of combinatorial data

Novel analysis methods for combinatorial data – data mining using machine learning

Incorporate existing databases (ICSD, etc.) into data analysis

Give input/feedback to theoretical calculations; AFLOWLIB, etc.

Database construction based on combinatorial data

Data mining of existing literature – data mining using machine reading

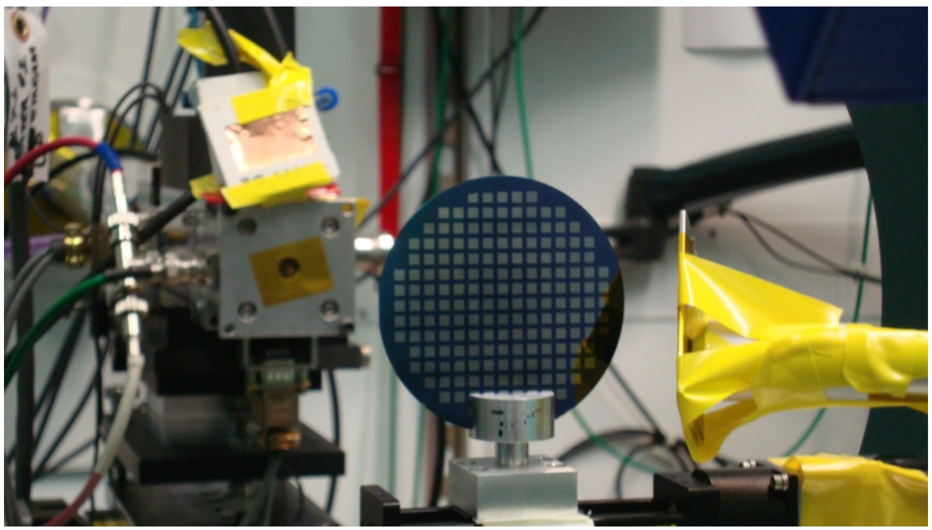
Goals: speed up discovery of new compounds and new relationships; curation of databases which can be used for future use: data mining, etc.

Rapid structural mapping of combinatorial wafers at synchrotron

Synchrotron diffraction set up at SSRL

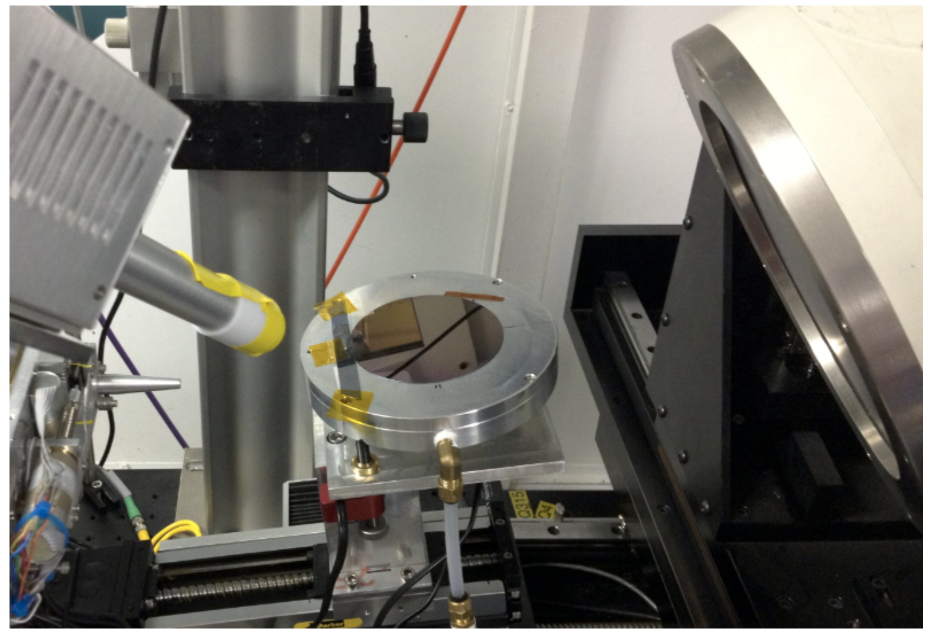
The entire 3” wafer (300 spots) can now be measured in 2 hrs

Transmission set up



XRF carried out simultaneously

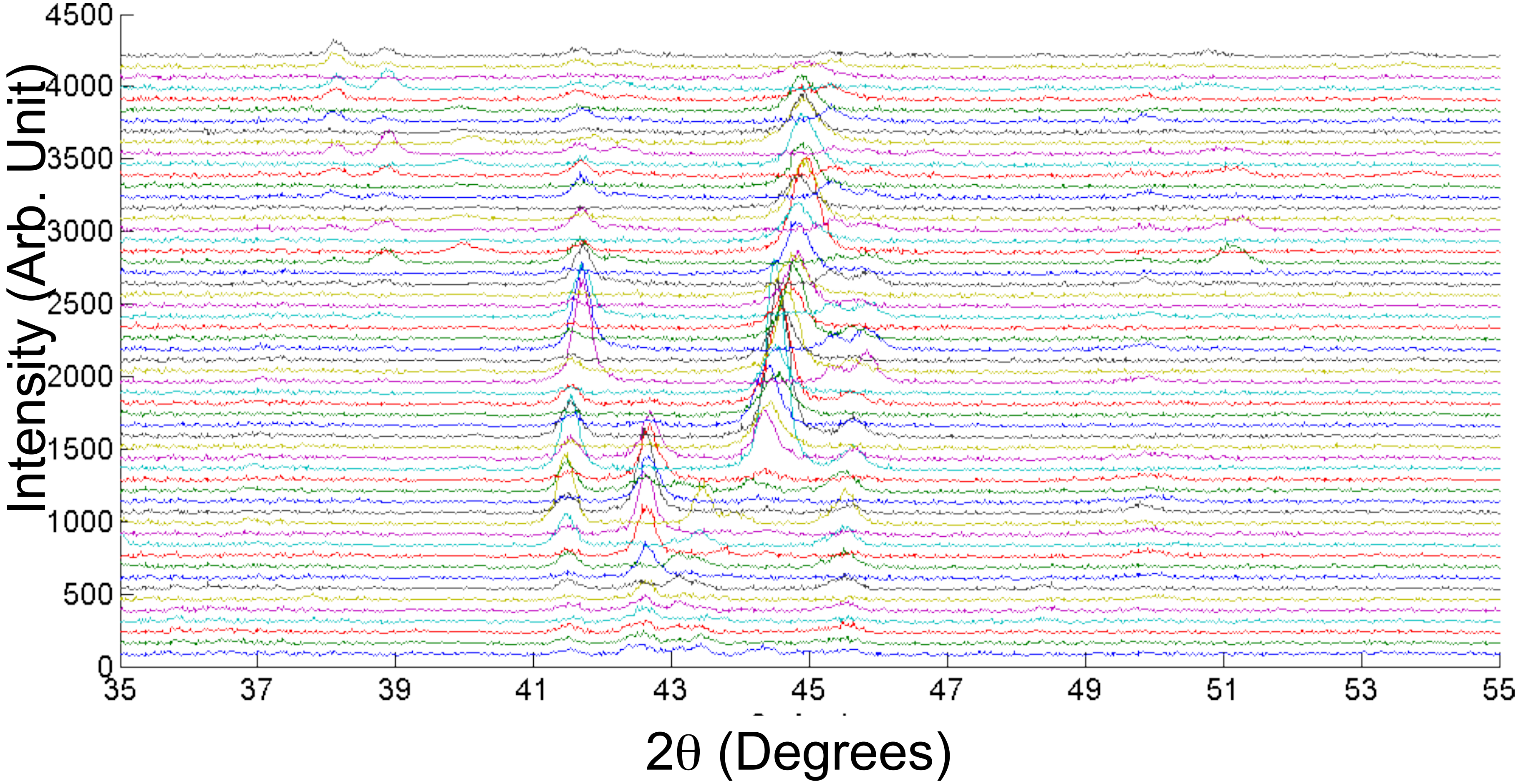
Reflection set up



Each wafer produces: 300 MB to 2 GB of image data

w/ A. Mehta

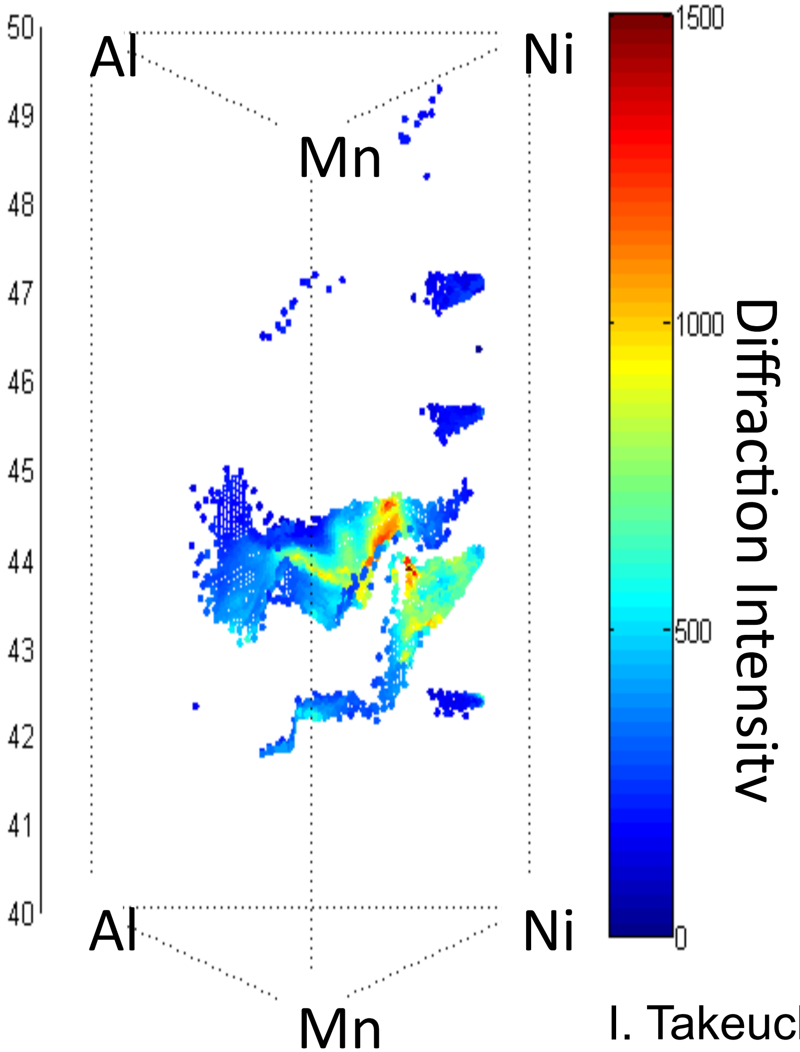
Hundreds of XRD Spectra are difficult to analyze by hand



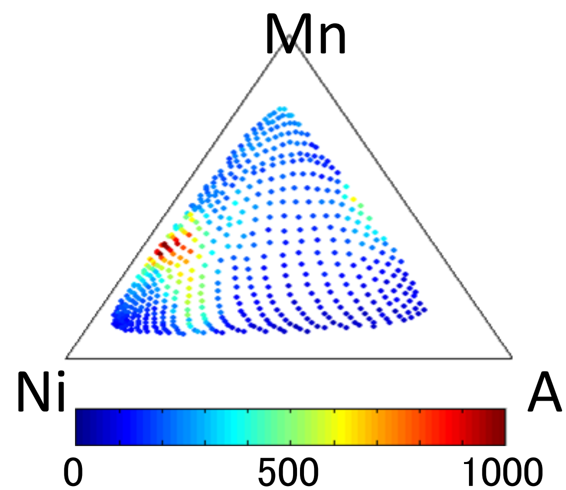
The same is true for any spectral data (Raman, FTIR, etc.)

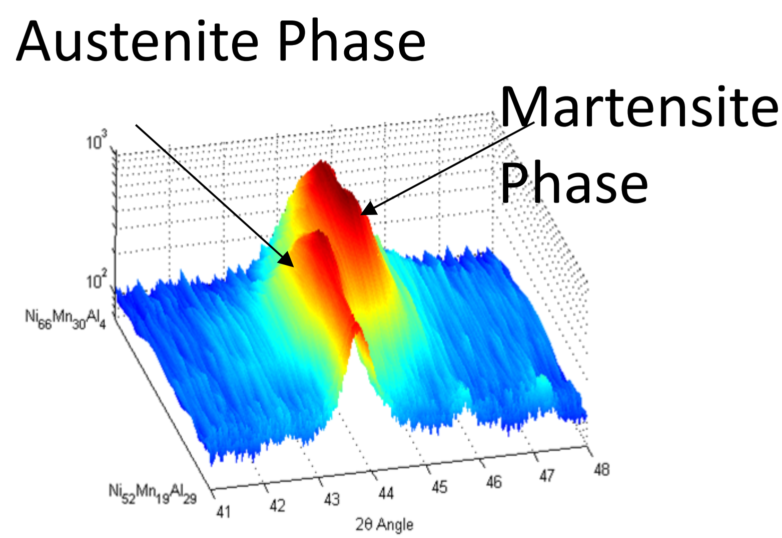
First step is visualization

Peak Angle and Intensity

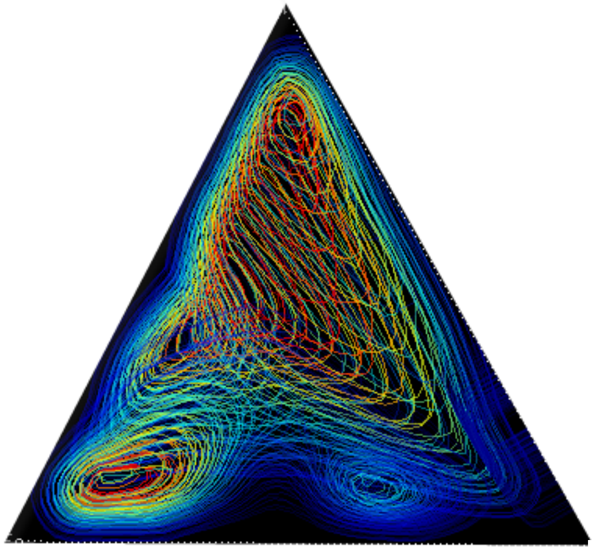


Intensity Cross Section





I. Takeuchi *et al.*, Rev. Sci. Instrum. 76, 062223 (2005)



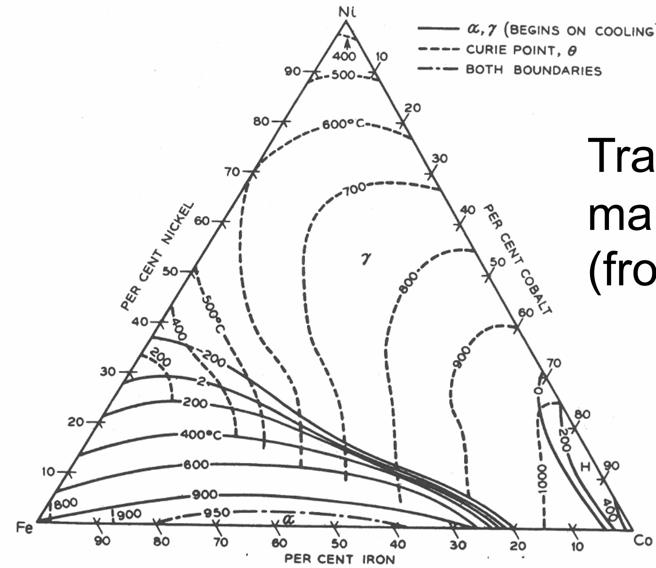


FIG. 5-77. The α,γ transitions and the Curie points observed during the cooling of Fe-Co-Ni alloys.

Transition mapping (from Bozarth)

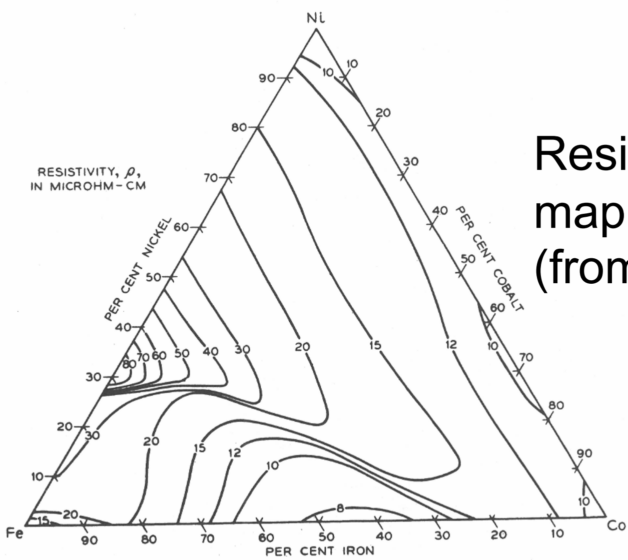


FIG. 5-78. Electrical resistivities of annealed Fe-Co-Ni alloys.

Resistivity mapping (from Bozarth)

Takeuchi *et al.*, MRS Bulletin **31**, 999 (2006)

Comparing XRD Spectra Quantitatively

Comparing Spectra Using the Pearson Correlation Coefficient:

$$r_{xy} = \sum_{i=1}^{n} \frac{(x_i - \overline{x})(y_i - \overline{y})}{\sigma_x \sigma_y}$$

***r**~xy~* **= 1 → Identical spectra**

r_{xy} **= 0 → No correlation**

r_{xy} **= -1 → Anti-correlation**

The spectra are either similar (i.e. they have the same structure) or they are not (i.e. different structures).

Therefore, we rescale the correlation coefficient to describe the dissimilarity of two spectra, instead of the correlation.

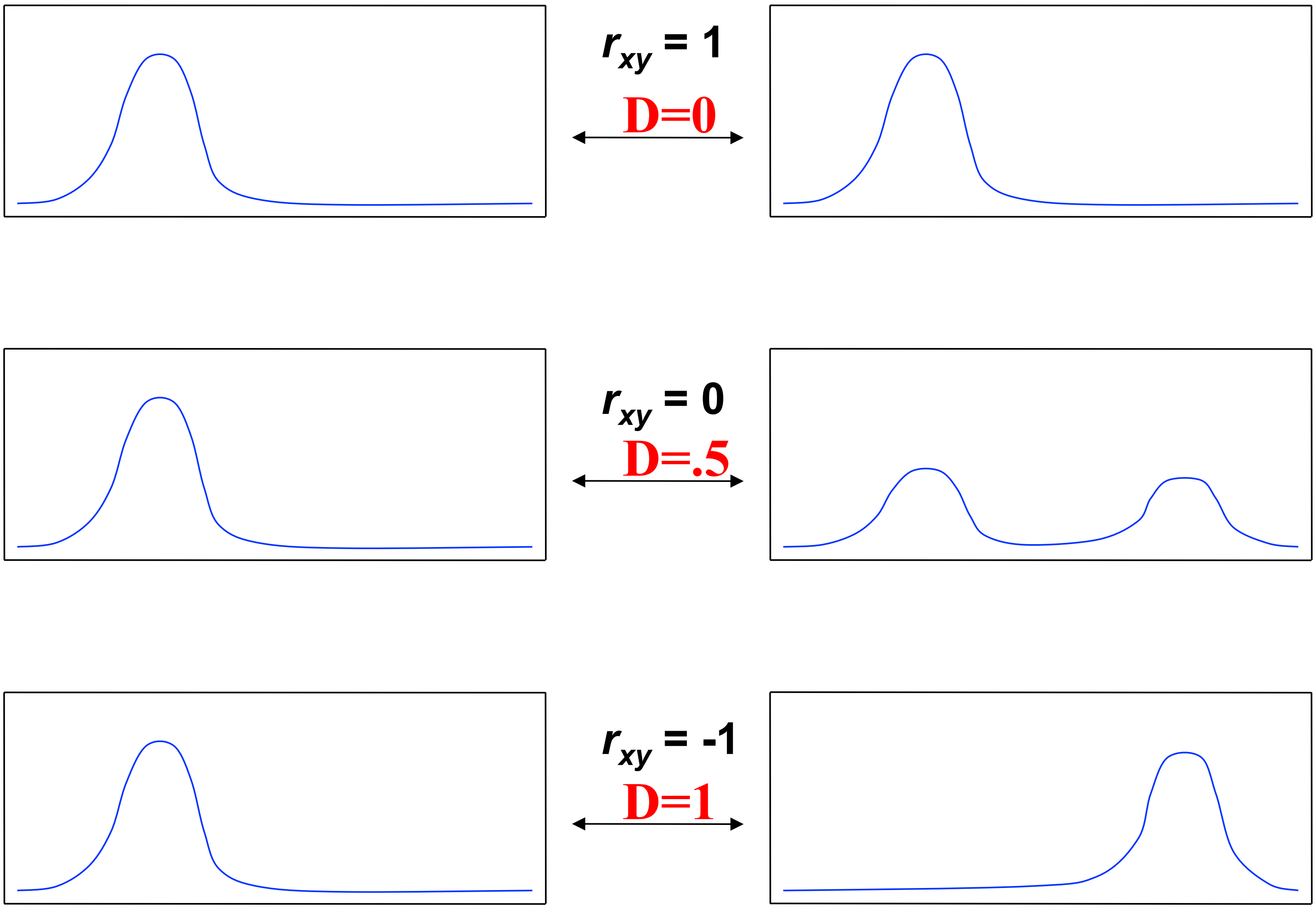
$$D_{xy} = (1 - r_{xy})/2$$

r_{xy} **= 1 → D = 0 → Identical spectra**

r_{xy} **= 0 → D = 1/2 → Dissimilar spectra**

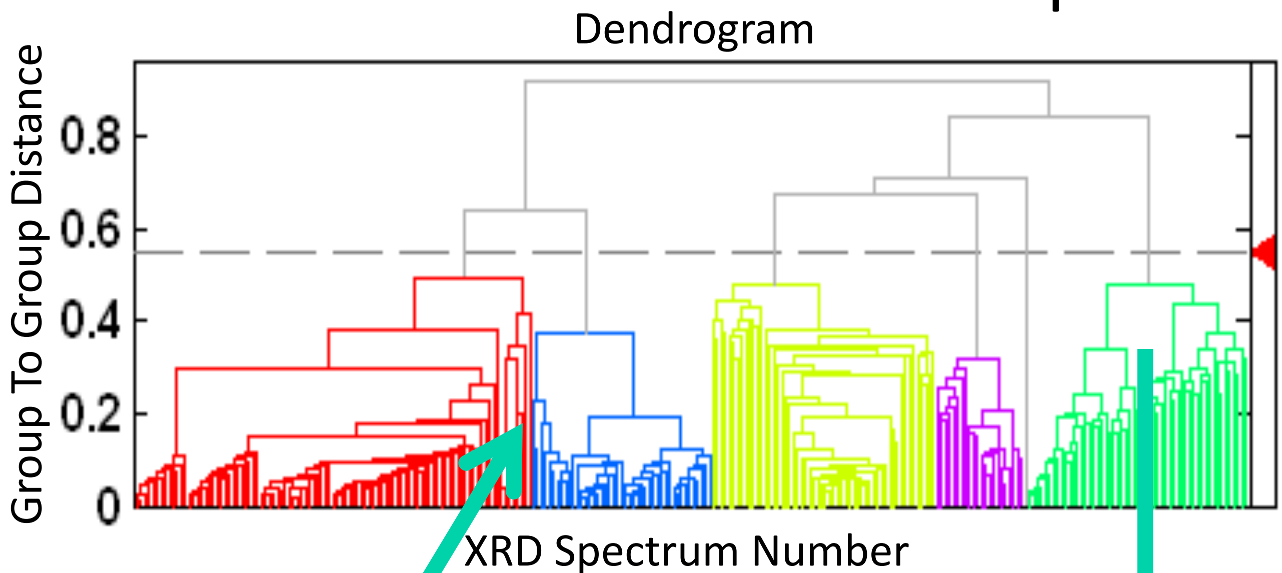
r_{xy} **= -1 → D = 1 → Very dissimilar spectra**

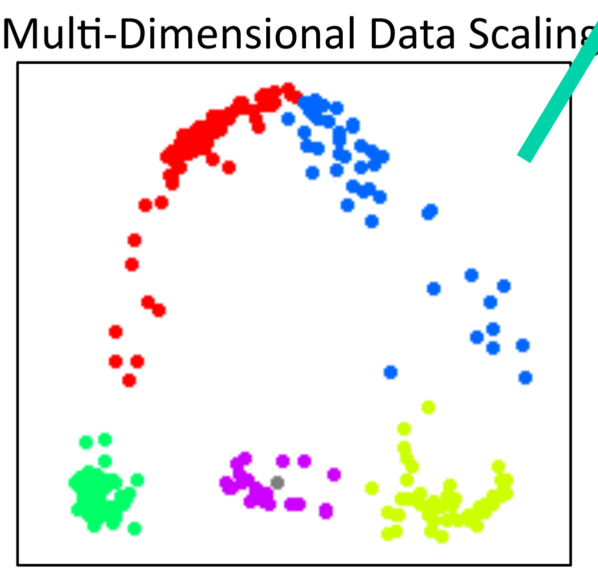
Comparing XRD spectra

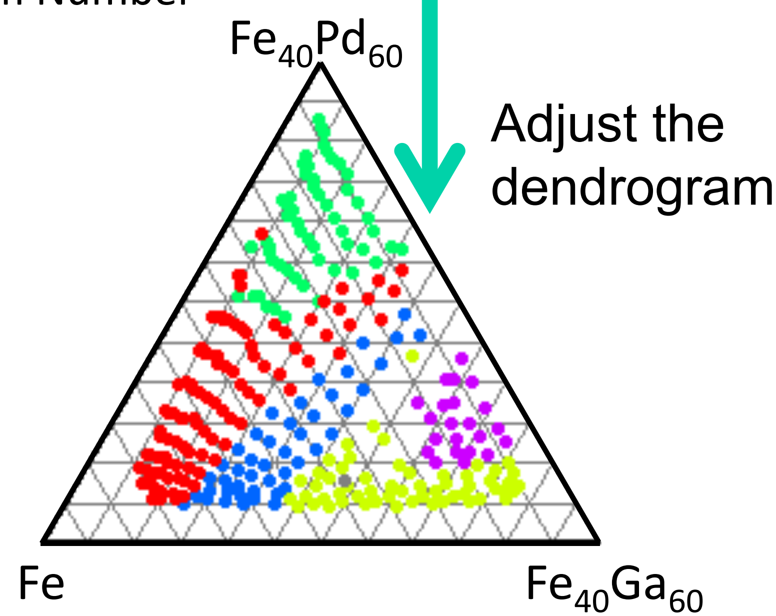


D = correlation coefficient

Analyze all spectra together using cluster analysis: Look for similarities between spectra





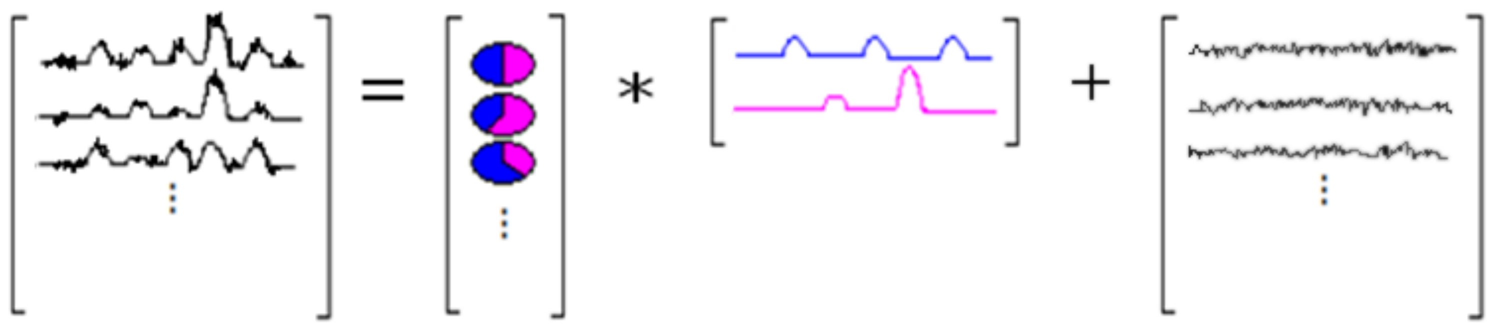


C. J. Long, et al. *Rev. Sci. Instrum*. 78, 072217 (2007)

Another analysis method:

Non-Negative Matrix Factorization (NMF):

(The Basic Idea)

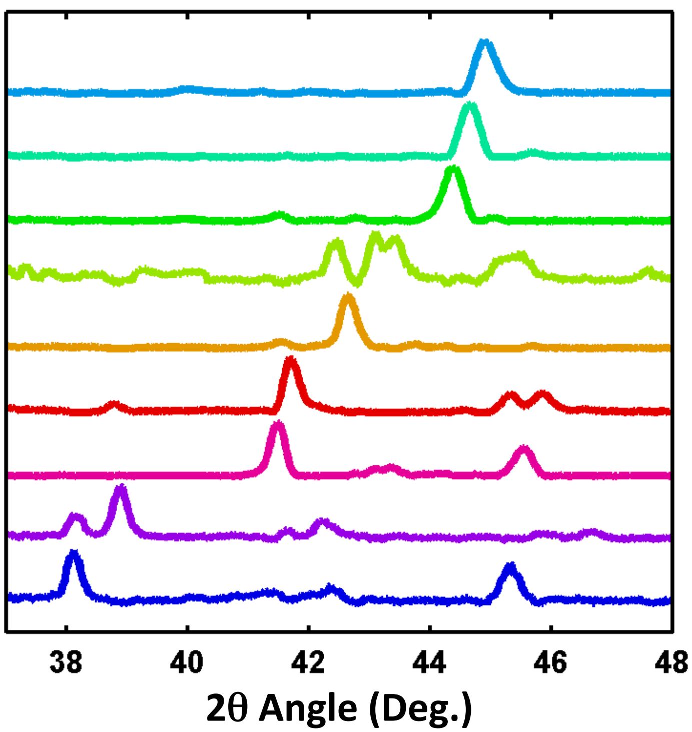


S	=	***W***	*	***B***	+	***E***
Experimental Spectra		**Weights**		**Basis Spectra**		**Residual Error**

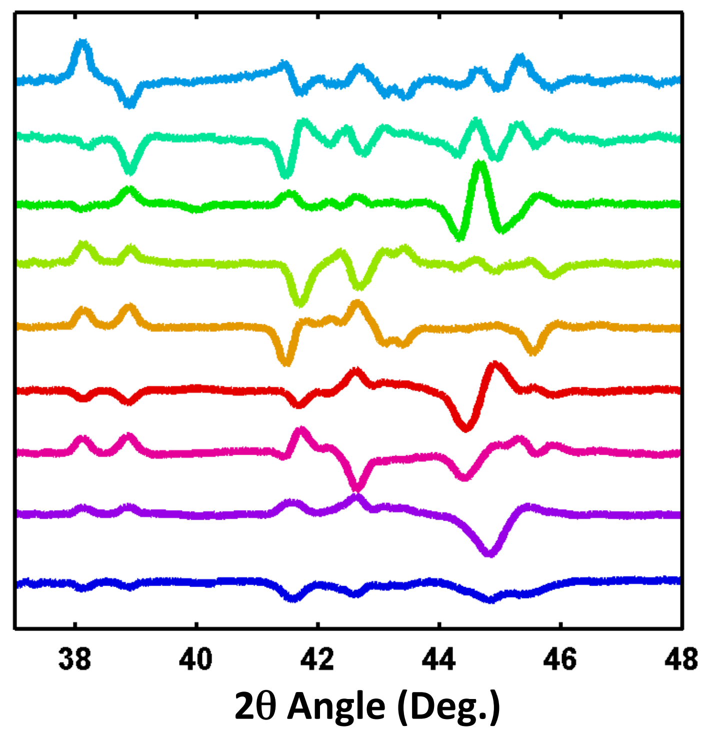
Experimental Spectra are Deconvolved

Comparison of NMF to PCA (principal component analysis)

Basis Patterns from NMF



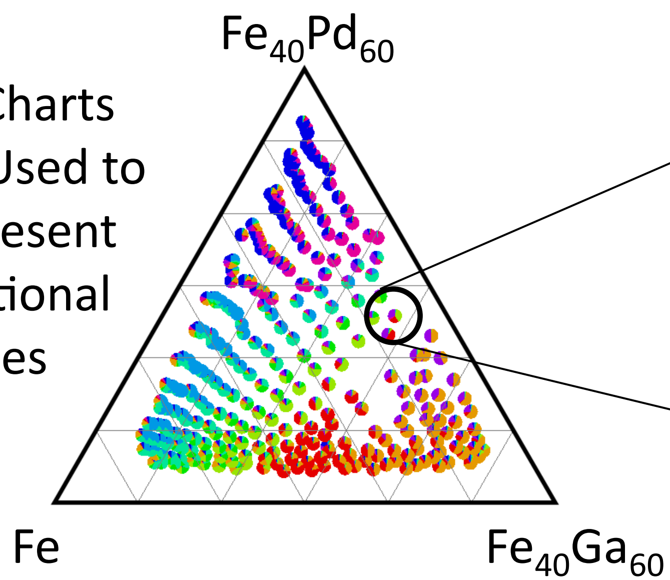
Basis Patterns from PCA

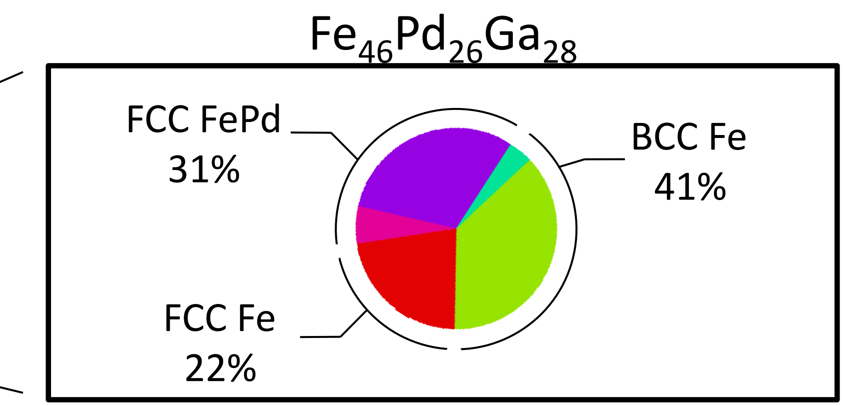


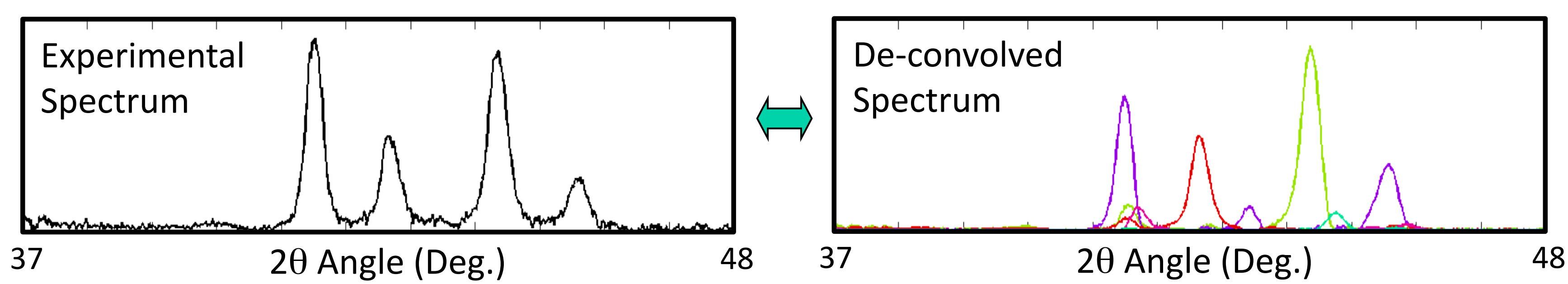
NMF produces basis patterns which look like diffraction spectra

Quantitative Identification of Mixed Phases

Pie Charts are Used to Represent Fractional Phases

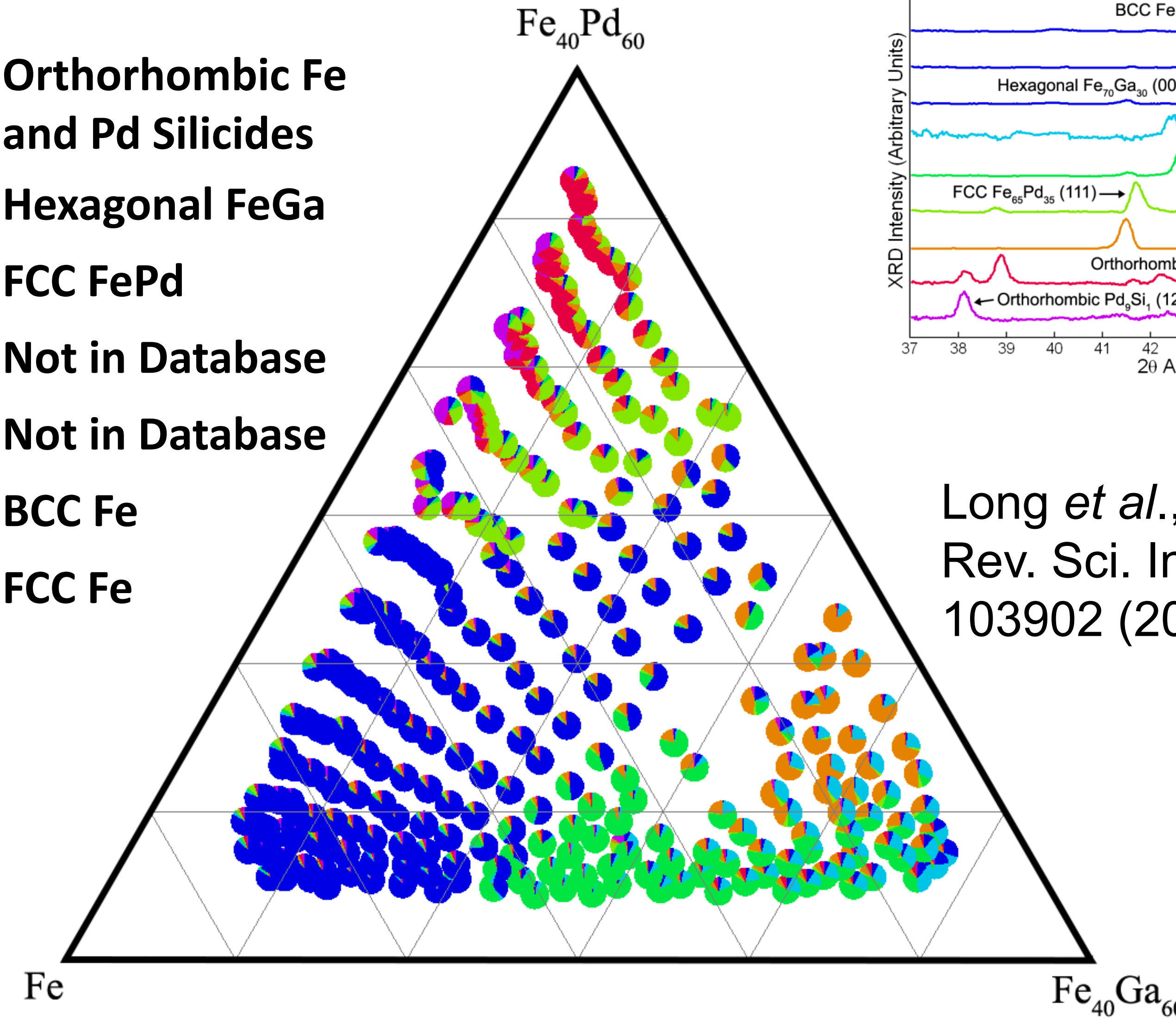


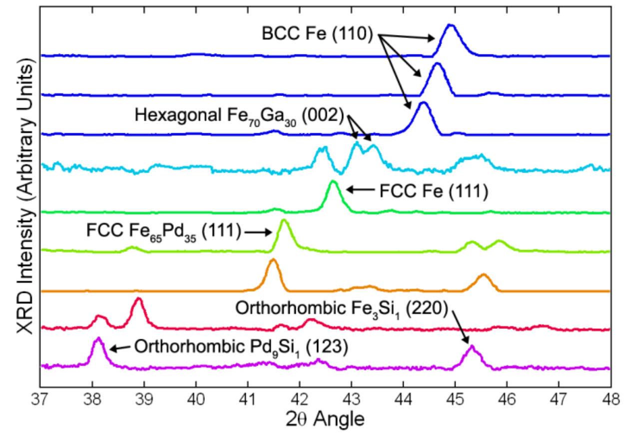




Working toward a structural phase diagram using NMF

- Orthorhombic Fe and Pd Silicides
- Hexagonal FeGa
- FCC FePd
- Not in Database
- Not in Database
- BCC Fe
- FCC Fe





Long *et al.*,
Rev. Sci. Instrum. **80**,
103902 (2009)

Inorganic Crystal Structure Database

- The world's largest database for completely identified inorganic crystal structures.
  - Around 150,000 critically evaluated material structure data entries.
    - 1,543 crystal structures of the elements.
    - 26,617 records for binary compounds.
    - 50,779 records for ternary compounds.
    - 51,118 records for quarternary and quintenary compounds.
    - About 105,000 entries (74,9%) have been assigned a structure type.
    - There are currently 6,250 structure prototypes.

http://www.fiz-karlsruhe.de/icsd.html

Integrating ICSD with combi XRD data

Most ternary phase diagrams are not known

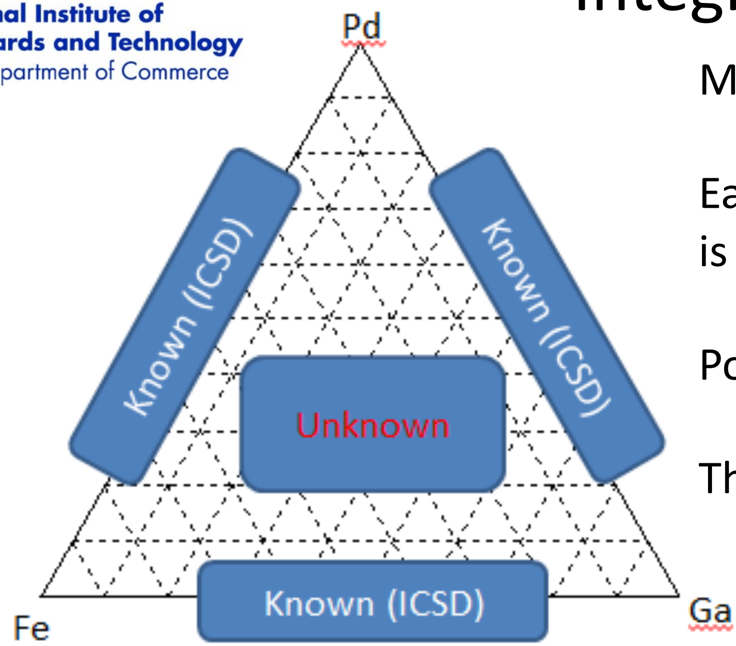
Each point on the ternary phase diagram
is one X-ray spectrum (expt)

Points on binary lines are simulated spectra from ICSD

They are rapidly mined/analyzed together

Kusne, et al.,
Scientific Reports **4**, 6367 (2014)

Integrating ICSD with combi XRD data



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mean shift theory

Kusne, et al.,
Scientific Reports **4**, 6367 (2014)

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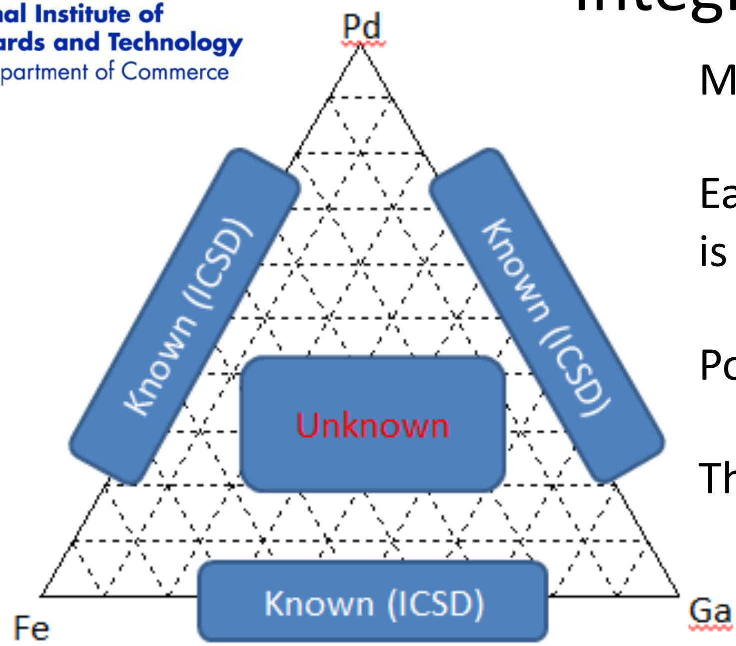

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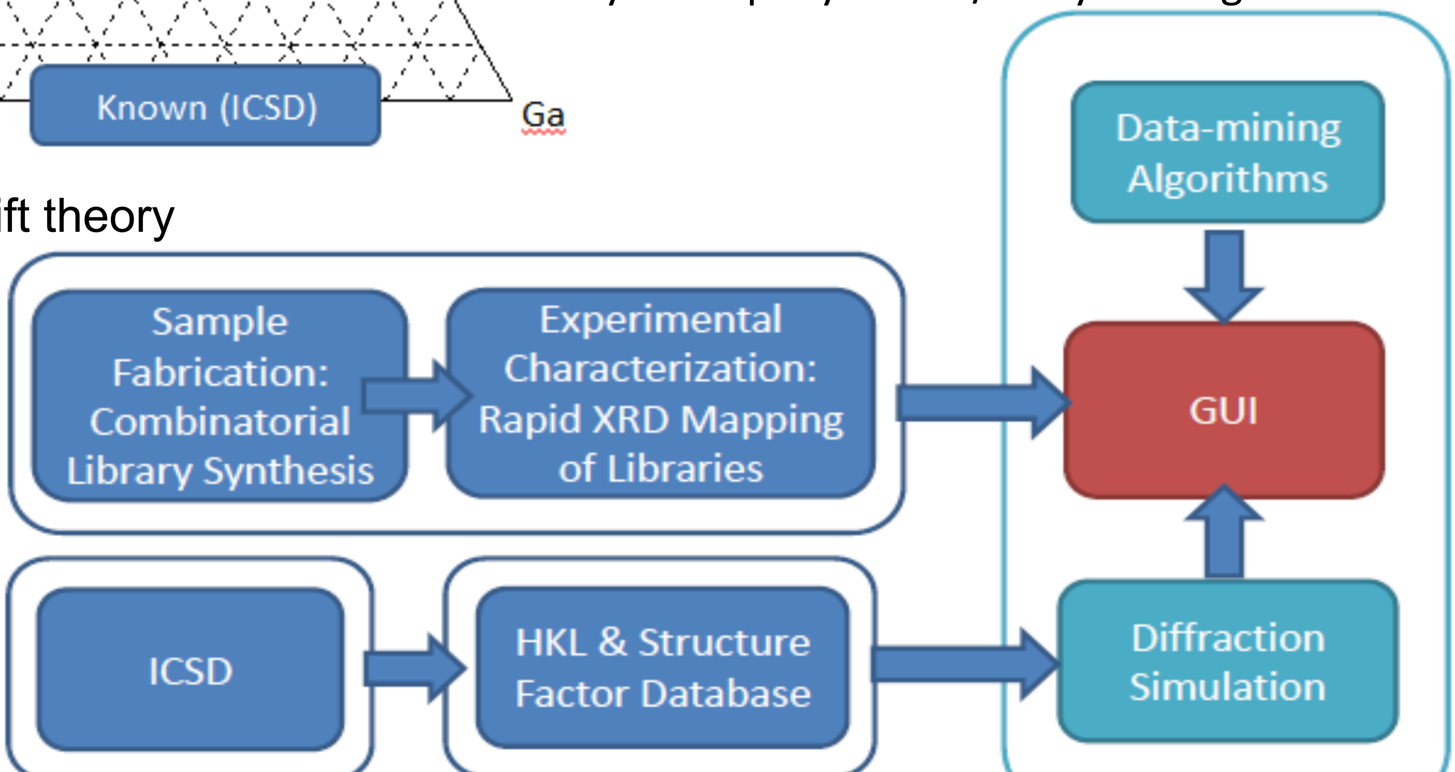

Scientific Reports **4**, 6367 (2014)

NIST
National Institute of Standards and Technology
U.S. Department of Commerce

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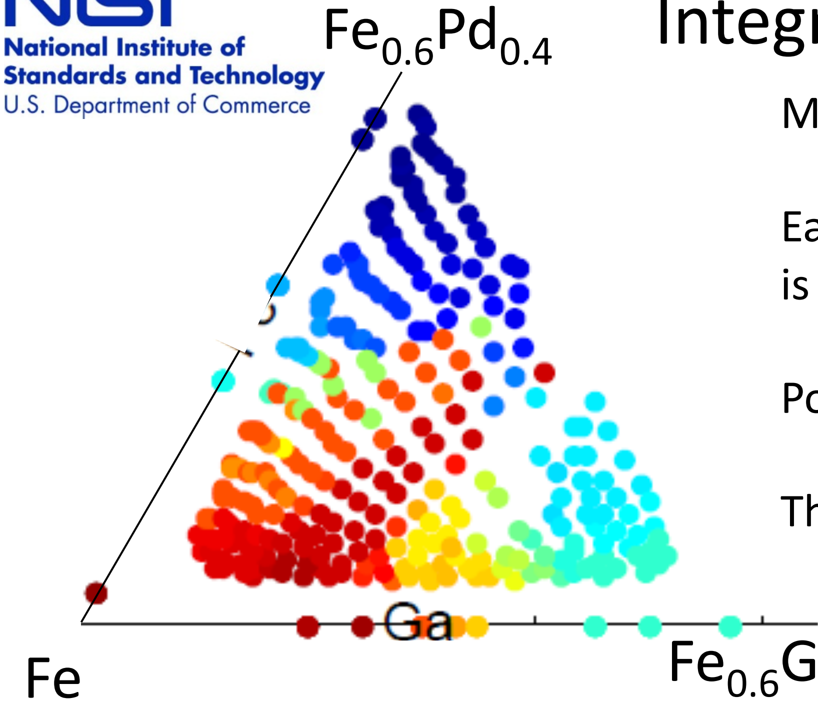
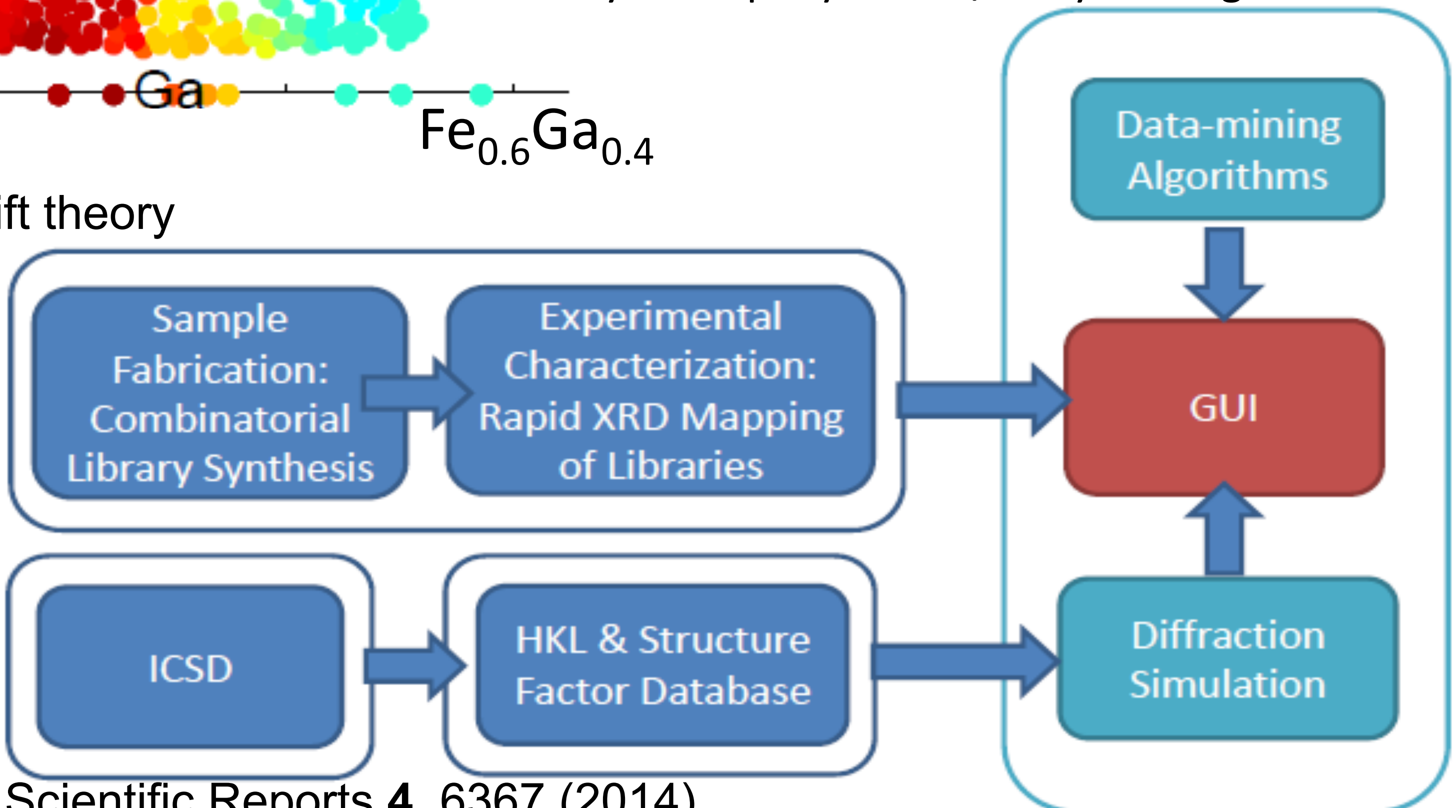


mean shift theory

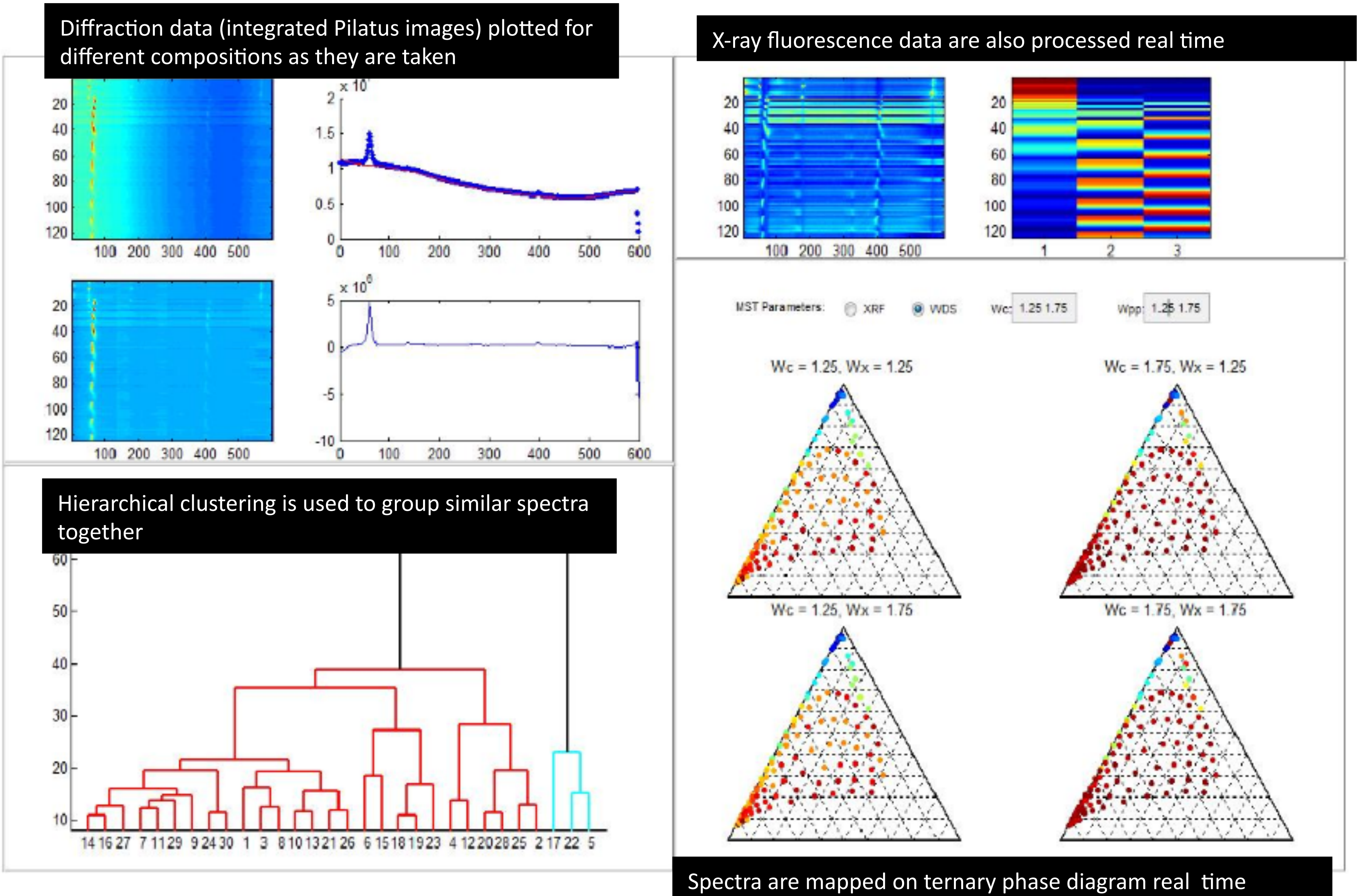


University of Maryland 1856

Scientific Reports **4**, 6367 (2014)

eV/atom

Real time analysis of combinatorial library data (synchrotron XRD); Integration with ICSD



Scientific Reports **4**, 6367 (2014)

Summary

Combinatorial experiments are the natural counterparts to computational efforts in the MGI

Handling large amount of data is always tricky, but we are beginning to see the light at the end of the tunnel

Machine learning techniques are actively being incorporated into analysis of combinatorial data

We are proposing to establish high- throughput experimentation centers/institutes