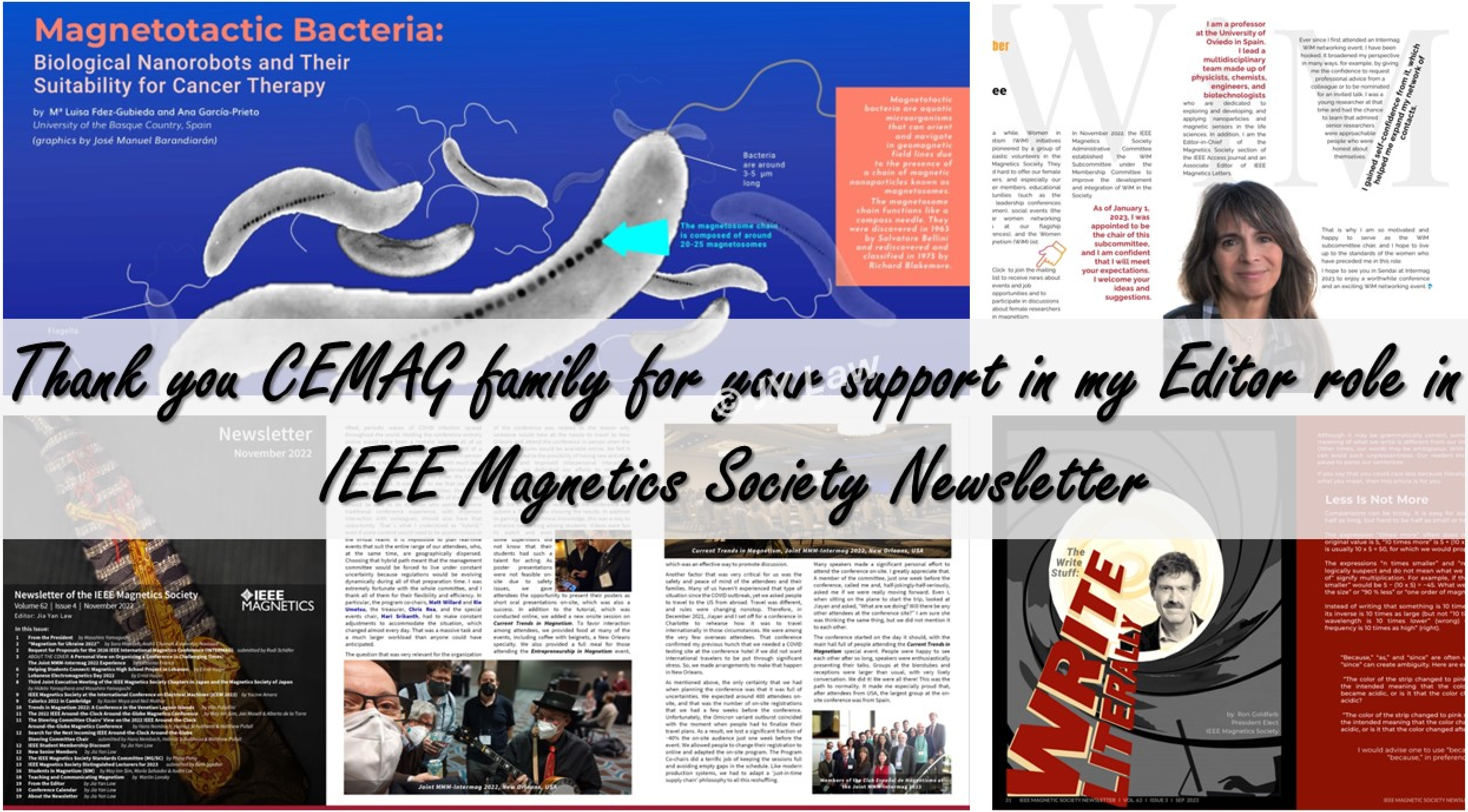
Magnetocaloric high-entropy alloys: prospects and challenges

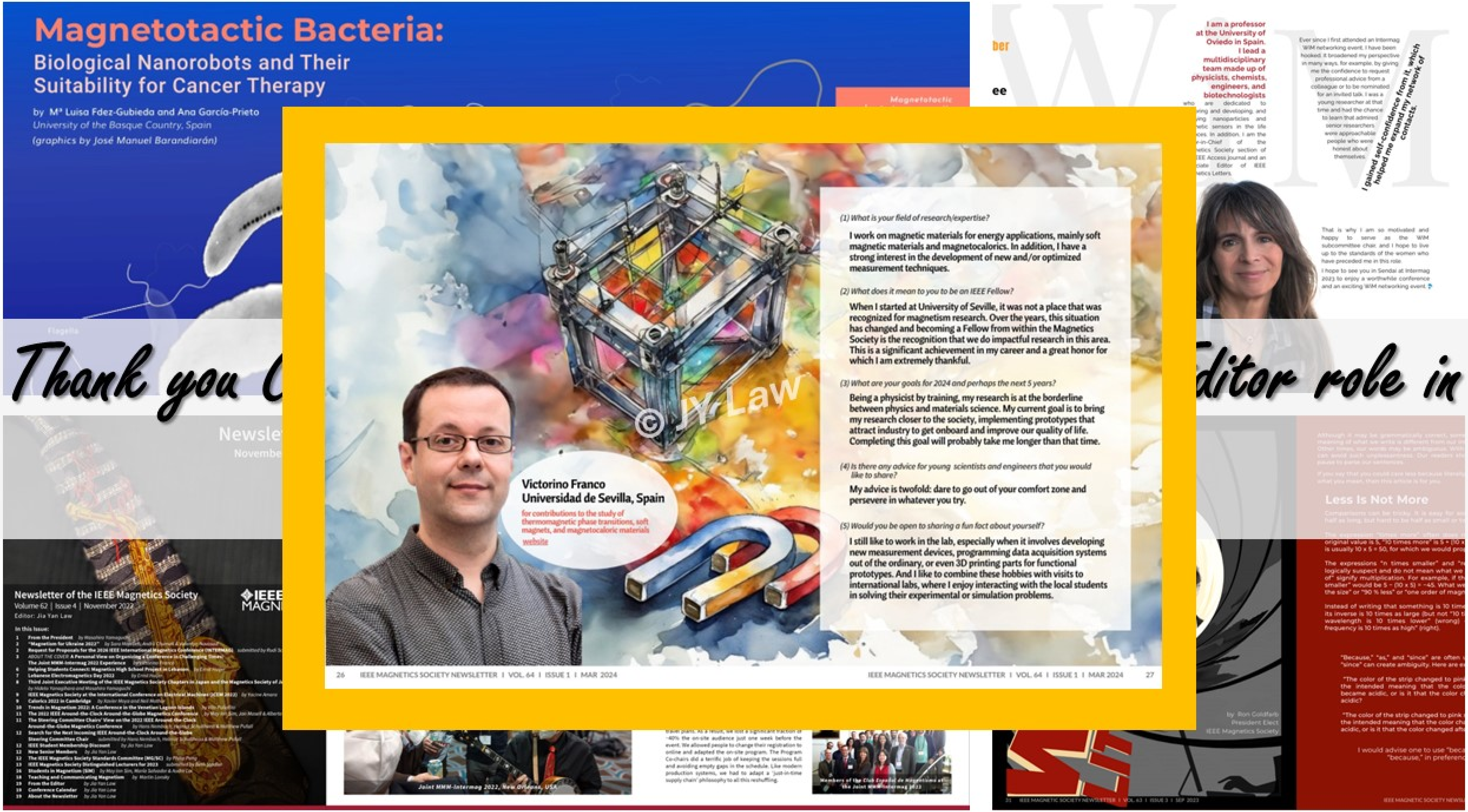
Jia Yan Law
University of Seville, Spain
jylaw@us.es

Thank you

CEMAG

Club Español de Magnetismo


Thank you CEMAG family for your support in my Editor role in IEEE Magnetics Society Newsletter
Magnetotactic Bacteria:
Biological Nanorobots and Their Suitability for Cancer Therapy
by Mª Luisa Fdez-Gubieda and Ana García-Prieto
University of the Basque Country, Spain
(graphics by José Manuel Barandiarán)
Bacteria are around 3-5 μm long
The magnetosome chain is composed of around 20-25 magnetosomes
Magnetotactic bacteria are aquatic microorganisms that can orient and navigate in geomagnetic field lines due to the presence of a chain of magnetic nanoparticles known as magnetosomes.
The magnetosome chain functions like a compass needle. They were discovered in 1963 by Salvatore Bellini and rediscovered and classified in 1975 by Richard Blakemore.
I am a professor at the University of Oviedo in Spain. I lead a multidisciplinary team made up of physicists, chemists, engineers, and biotechnologists
As of January 1, 2023, I was appointed to be the chair of this subcommittee, and I am confident that I will meet your expectations. I welcome your ideas and suggestions.
I gained self-confidence from it, which helped me expand my network of contacts.
Newsletter
November 2022
Newsletter of the IEEE Magnetics Society
Volume 62 | Issue 4 | November 2022
Editor: Jia Yan Law
IEEE MAGNETICS
Current Trends in Magnetism, Joint MMM-Intermag 2022, New Orleans, USA
Joint MMM-Intermag 2022, New Orleans, USA
Less Is Not More
The Write Stuff:
WRITE LITERALLY
by Ron Goldfarb
President Elect
IEEE Magnetics Society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Thank you
Editor role in
(1) What is your field of research/expertise?
I work on magnetic materials for energy applications, mainly soft magnetic materials and magnetocalorics. In addition, I have a strong interest in the development of new and/or optimized measurement techniques.
(2) What does it mean to you to be an IEEE Fellow?
When I started at University of Seville, it was not a place that was recognized for magnetism research. Over the years, this situation has changed and becoming a Fellow from within the Magnetics Society is the recognition that we do impactful research in this area. This is a significant achievement in my career and a great honor for which I am extremely thankful.
(3) What are your goals for 2024 and perhaps the next 5 years?
Being a physicist by training, my research is at the borderline between physics and materials science. My current goal is to bring my research closer to the society, implementing prototypes that attract industry to get onboard and improve our quality of life. Completing this goal will probably take me longer than that time.
(4) Is there any advice for young scientists and engineers that you would like to share?
My advice is twofold: dare to go out of your comfort zone and persevere in whatever you try.
(5) Would you be open to sharing a fun fact about yourself?
I still like to work in the lab, especially when it involves developing new measurement devices, programming data acquisition systems out of the ordinary, or even 3D printing parts for functional prototypes. And I like to combine these hobbies with visits to international labs, where I enjoy interacting with the local students in solving their experimental or simulation problems.
Victorino Franco
Universidad de Sevilla, Spain
for contributions to the study of thermomagnetic phase transitions, soft magnets, and magnetocaloric materials
website
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26 IEEE MAGNETICS SOCIETY NEWSLETTER | VOL. 64 | ISSUE 1 | MAR 2024
IEEE MAGNETICS SOCIETY NEWSLETTER | VOL. 64 | ISSUE 1 | MAR 2024 27
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IEEE Magnetics Society
Joint MMM-Intermag 2022, New Orleans, USA

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University of Seville, Spain

Prof. Victorino Franco

Dr. Carlos Romero-Muñiz

Mr. Zhonghao Chang

Ms. Gloria Guerrero-Muñoz

Dr. Luis M. Moreno-Ramírez

Mr. Zhe Cui

Dr. Carmen González Galán

Mr. Aun N. Khan

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Ms. Elisa Guisado-Arenas

Magnetocaloric high-entropy alloys: prospects and challenges

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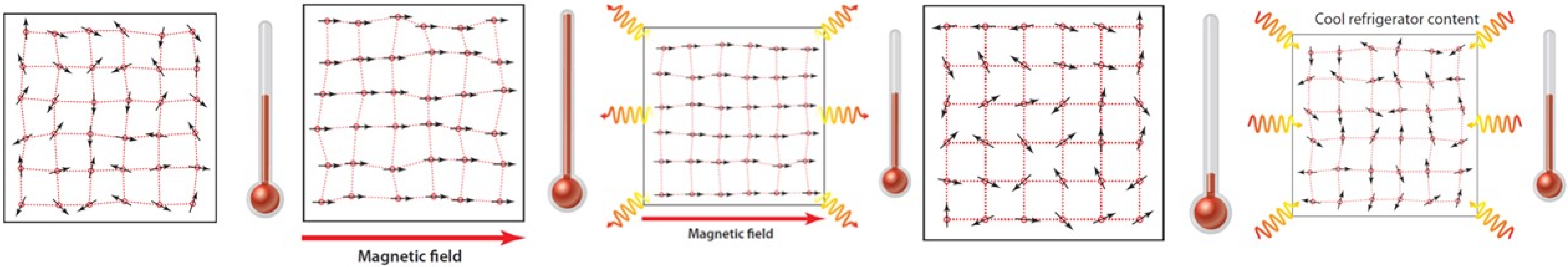
magnetocaloric effect

refers to **adiabatic** **ΔT** of magnetic material

when subjected to **varying** magnetic field

4 stages of

Magnetic Refrigeration Cycle



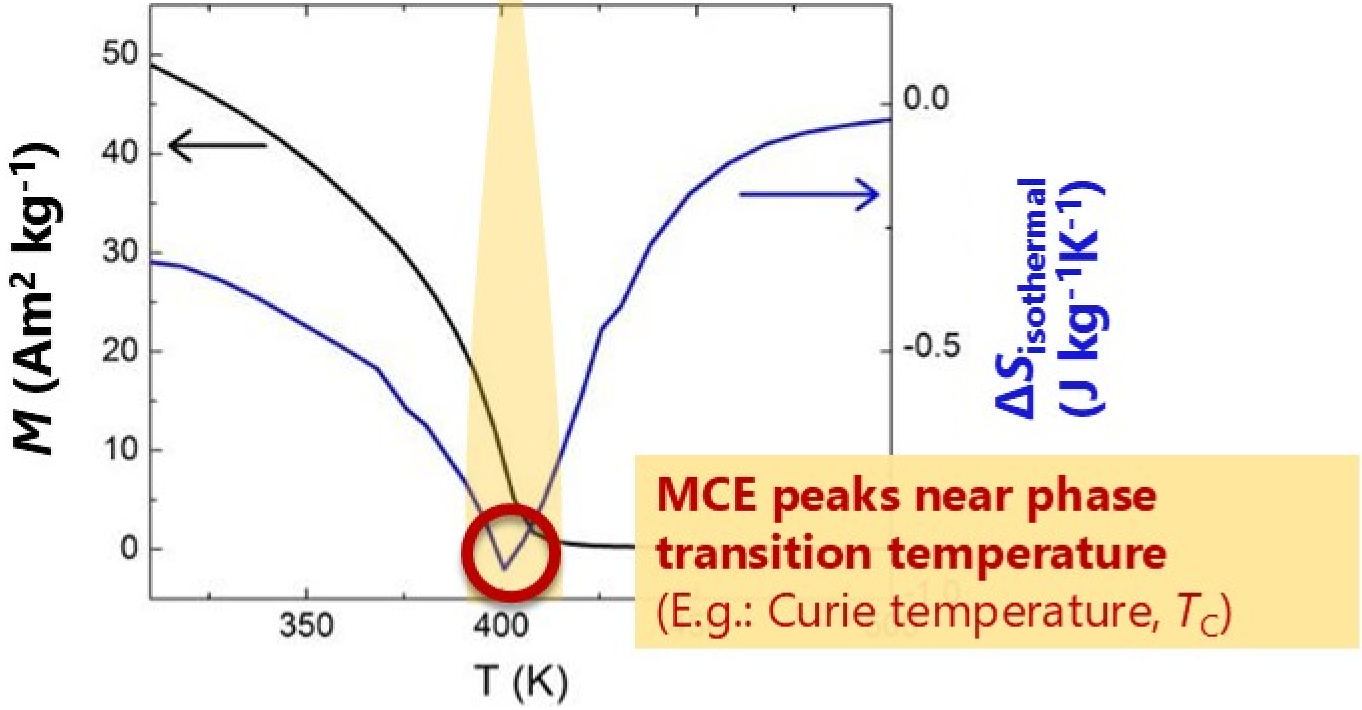
Adiabatic magnetization | Heat removal | Adiabatic demagnetization | Load cooling

Magnetocaloric effect ≡ ΔT or $\Delta S_{isothermal}$

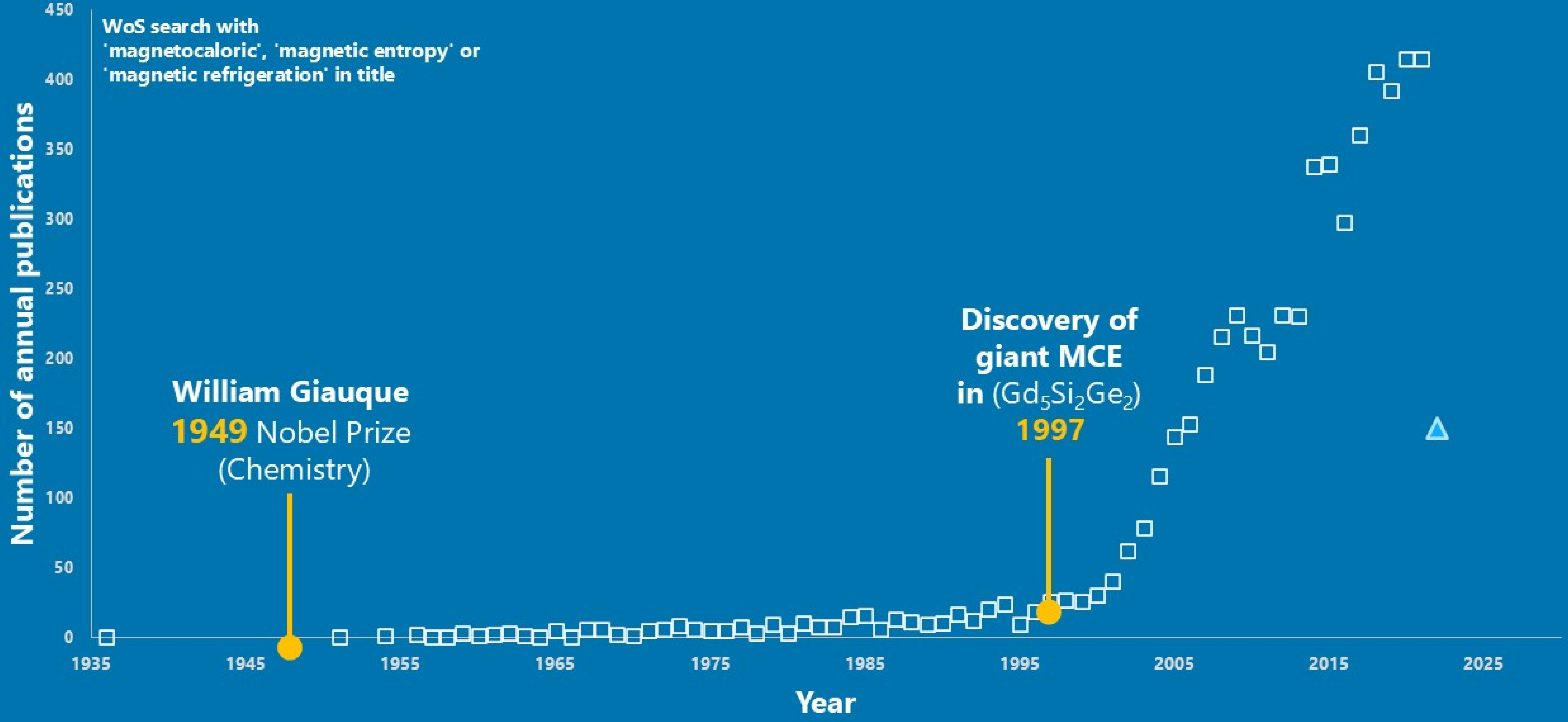
Prog. Mater. Sci. 93 (2018) 112-232
MCE book, in preparation

$\Delta S_{isothermal}$ commonly reported using the magnetic measurement data…

$$|\Delta S_{isothermal}| = \mu_0 \int_0^{H_f} \left(\frac{\partial M}{\partial T}\right)_H dH$$

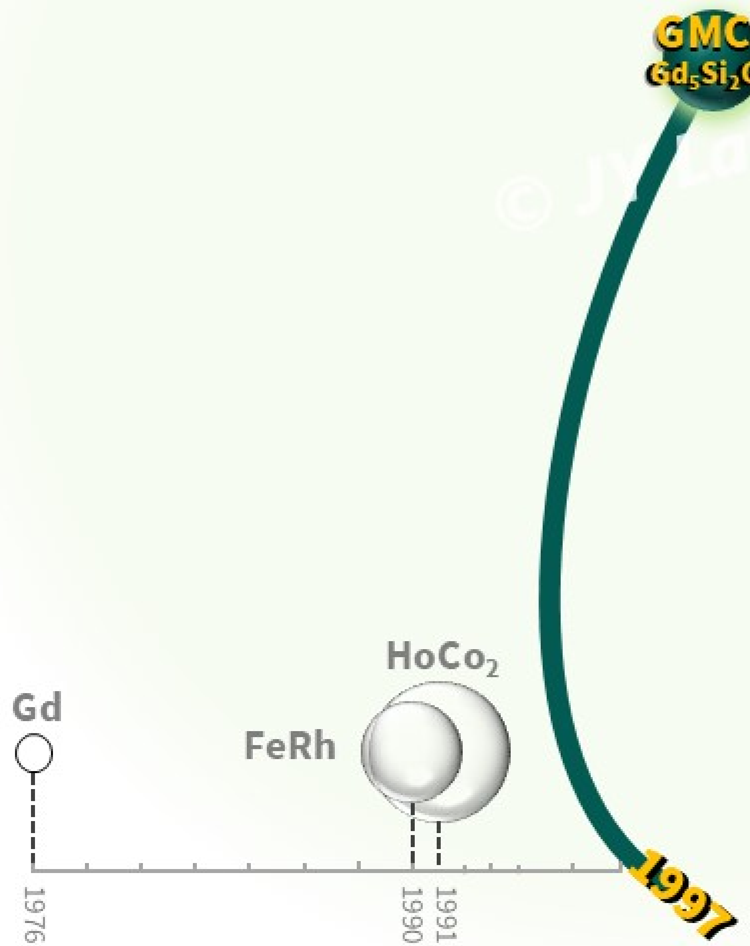


milestones



WoS search with
'magnetocaloric', 'magnetic entropy' or
'magnetic refrigeration' in title
William Giauque
1949 Nobel Prize
(Chemistry)
Discovery of
giant MCE
in ($Gd_5Si_2Ge_2$)
1997
Number of annual publications
Year
0
50
100
150
200
250
300
350
400
450
1935
1945
1955
1965
1975
1985
1995
2005
2015
2025

FOMT compounds that *flourished* after GMCE $Gd_5Si_2Ge_2$

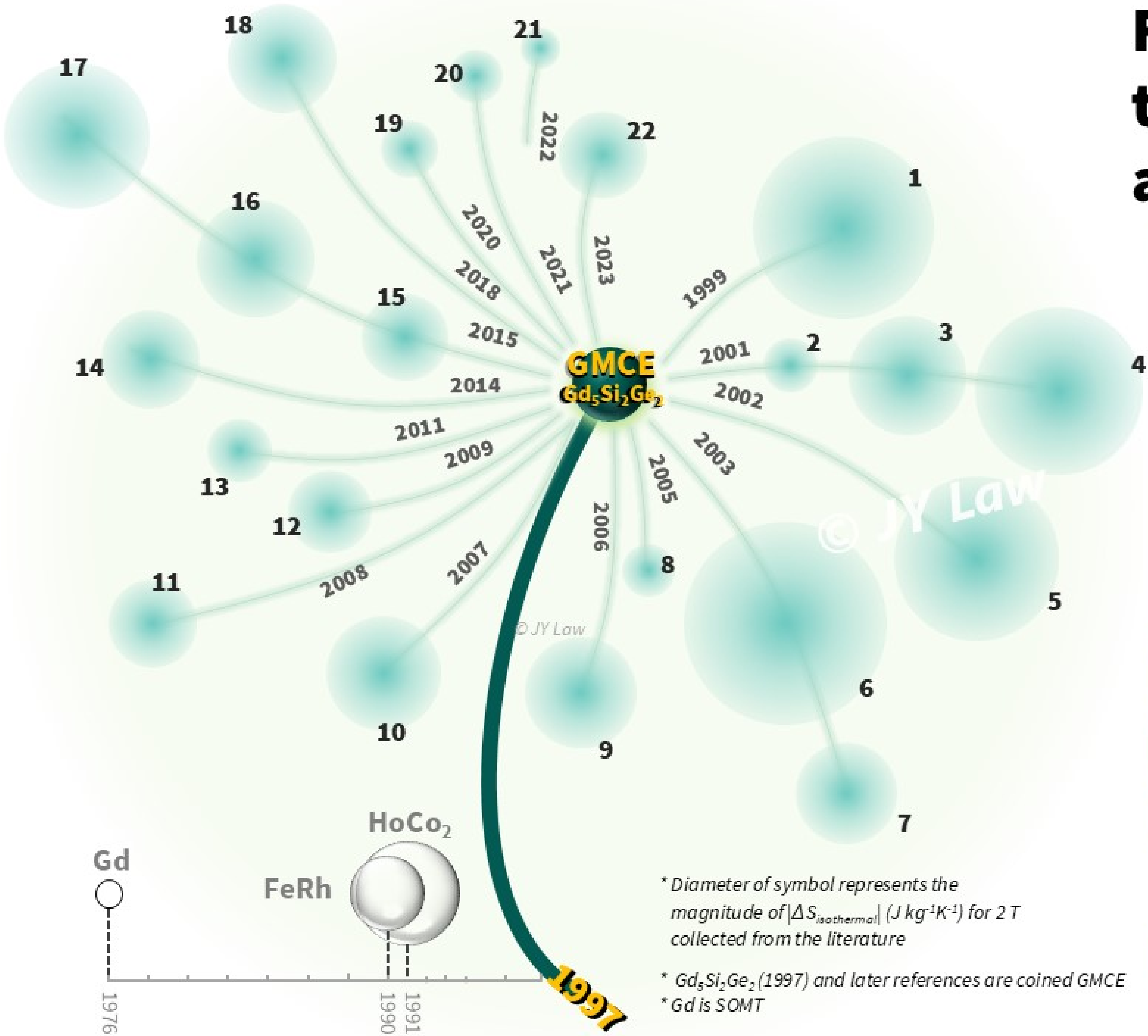


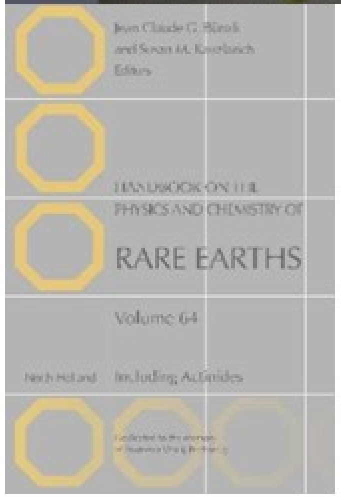
** Diameter of symbol represents the magnitude of $|\Delta S_{isothermal}|$ (J kg^{-1}K^{-1}) for 2 T collected from the literature*

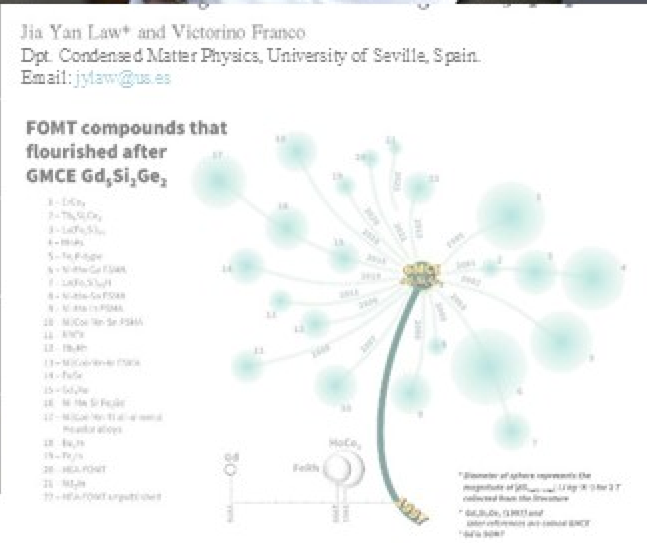
** $Gd_5Si_2Ge_2$ (1997) and later references are coined GMCE*

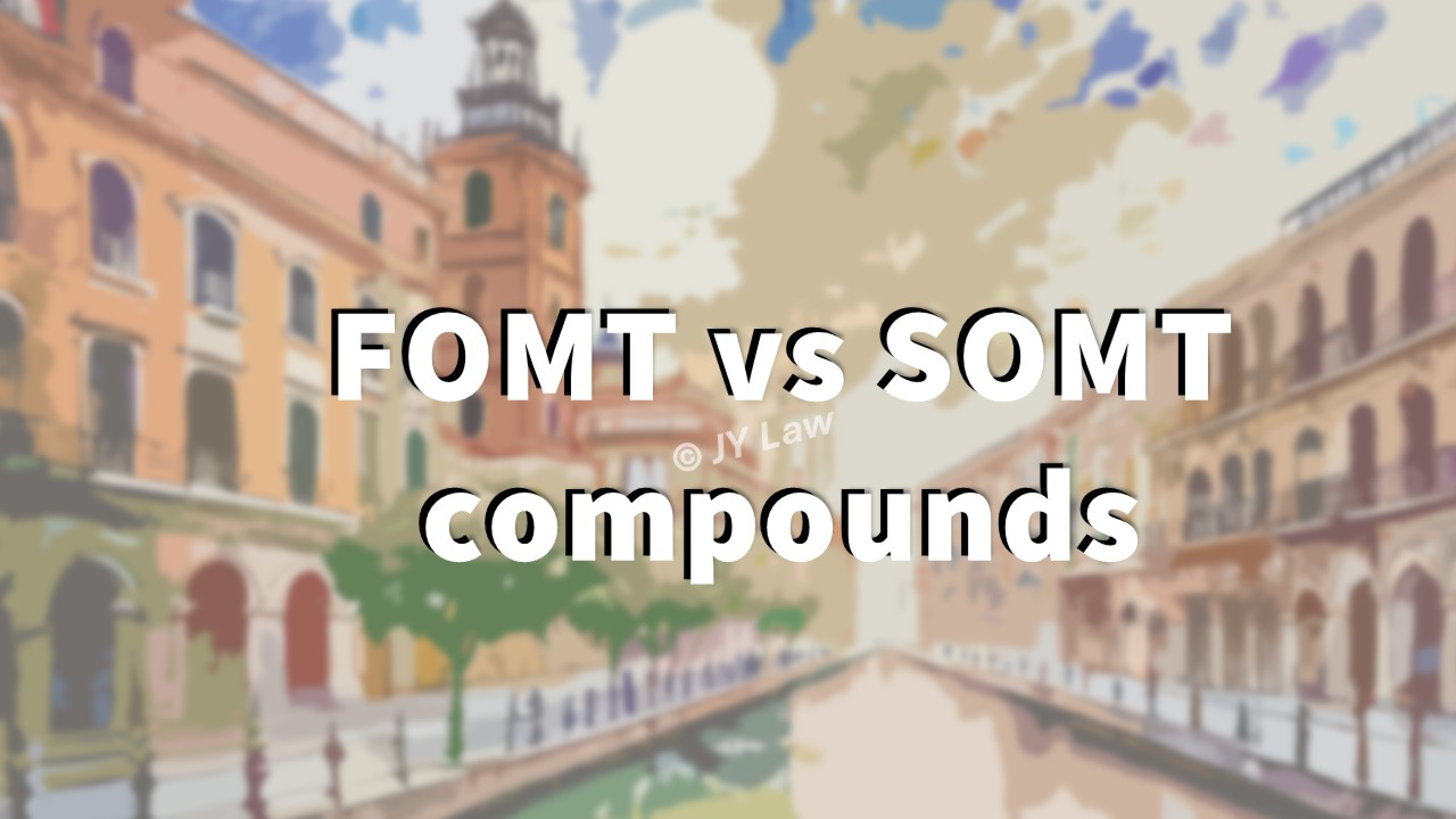
** Gd is SOMT*

FOMT compounds that *flourished* after GMCE $Gd_5Si_2Ge_2$








FOMT vs SOMT
compounds
© JY Law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• Moderate ΔS_{iso}
• wider T_{span}
• No thermal hysteresis
Large ΔS_{iso}
No thermal hysteresis
© JY Law
1st order
MCE
Larger ΔS_{iso} •
Narrow T_{span} •
Hysteresis •
Rate-dependent behavior •
2nd order

Advanced Energy Materials
Perspectives and energy applications of magnetocaloric, pyromagnetic, electrocaloric, and pyroelectric materials

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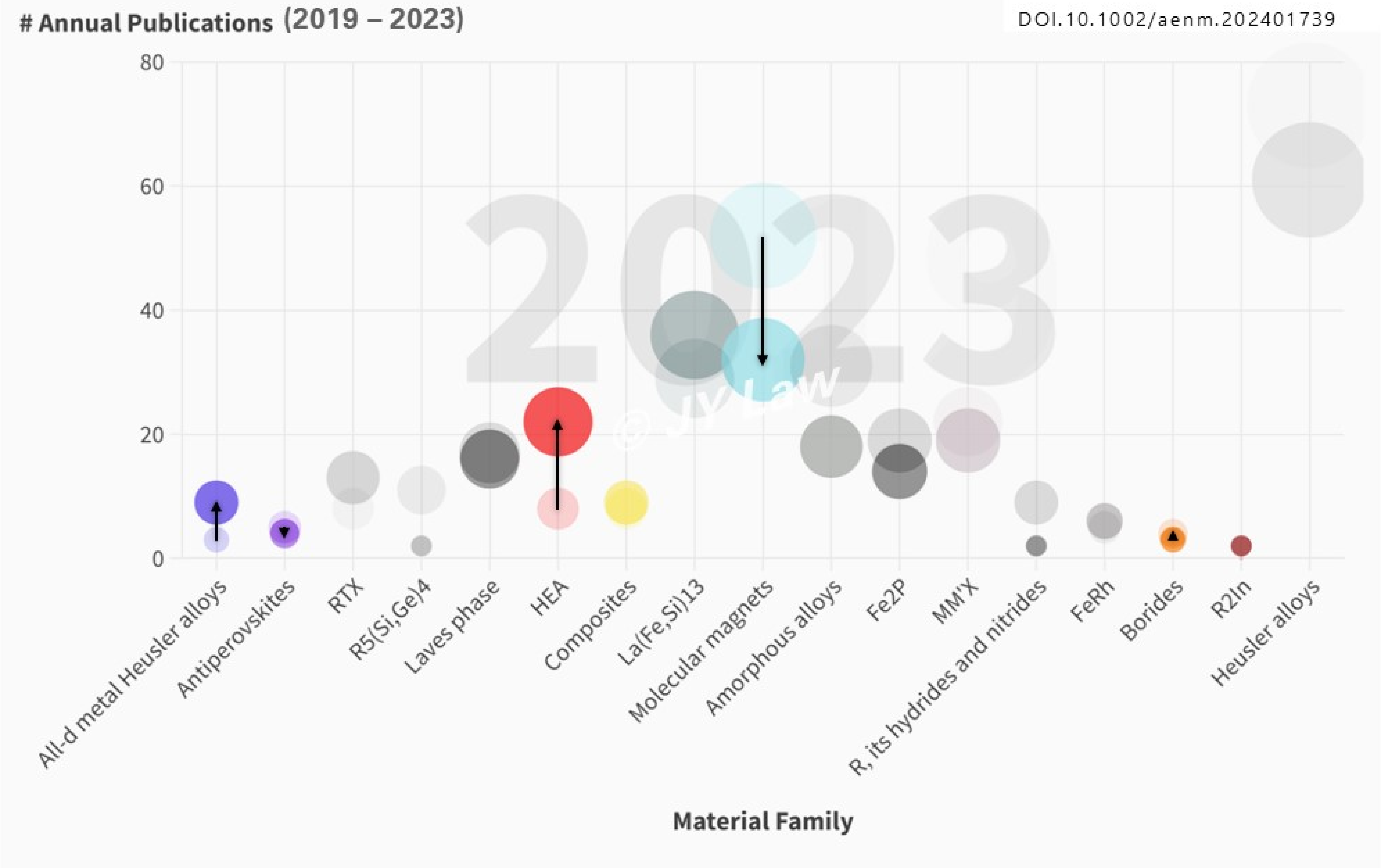
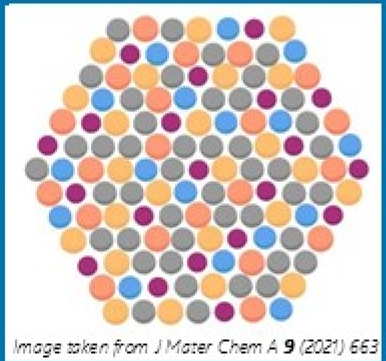
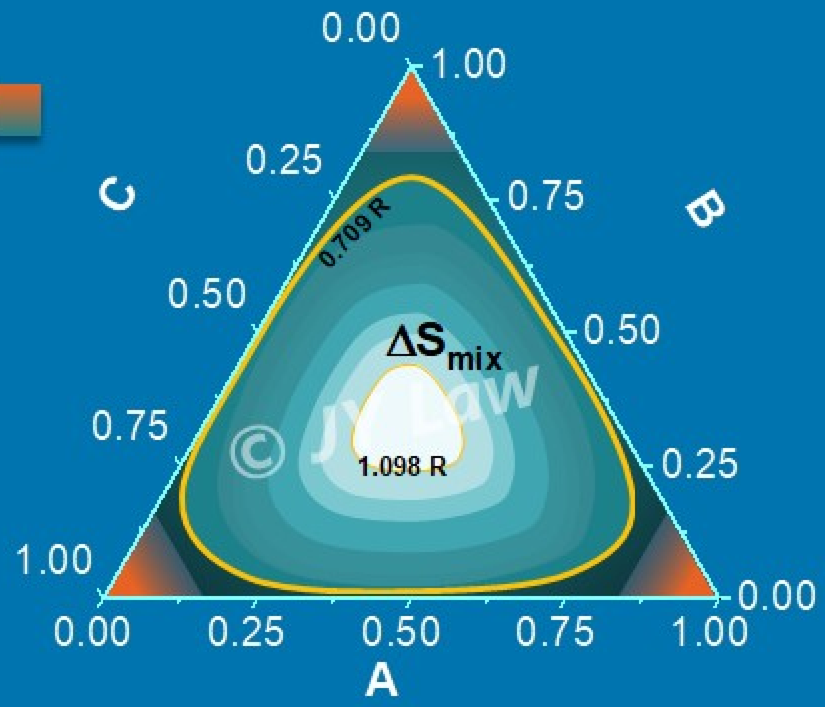


Concept of HEA

Traditional alloys or LEA

Equiatomic HEA





Contour plot of ΔS_{mix} of a model ternary alloy system

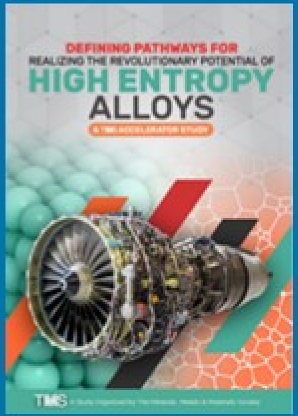
1^{st} generation HEAs:

restricted to

- ≥ 5 principal elements
- equiatomic compositions
- single-phase metallic solid solutions

Today

HEAs have evolved to 2^{nd} generation...



Concept of HEA

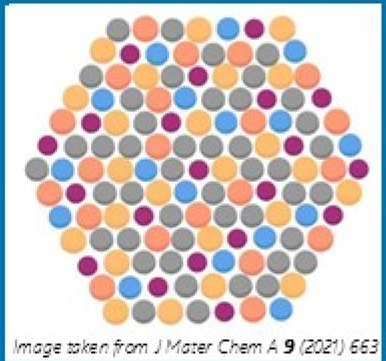
Configurational entropy of mixing

$$\Delta S_{mix} = -R \sum_{i=1}^{n} c_i \ln c_i$$

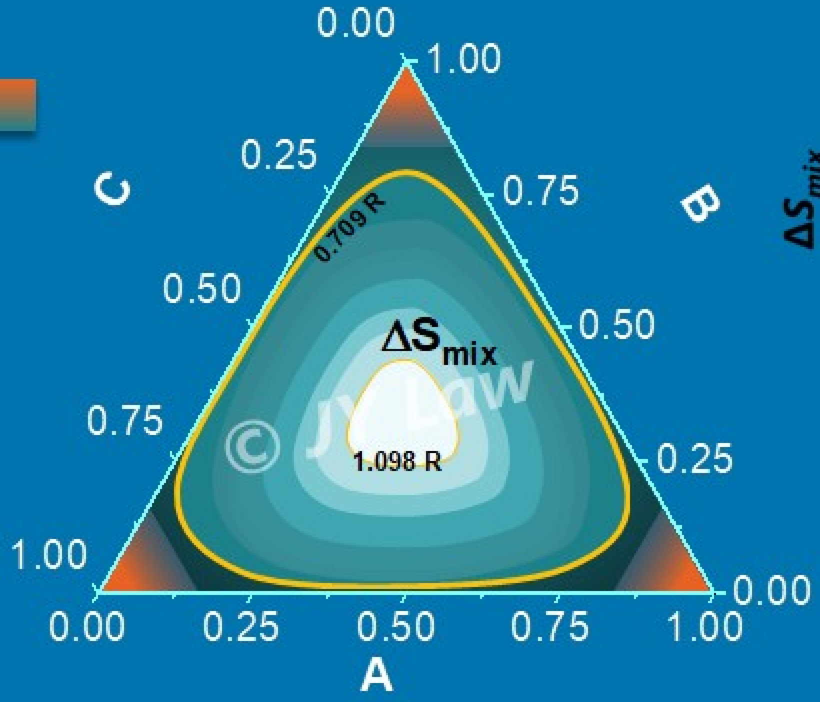
R = gas constant (8.314 J mol^{-1} K^{-1})
n = total # constituent elements
c_i = molar fraction of i^{th} element

Traditional alloys or LEA

Equiatomic HEA



*multi*principal elements



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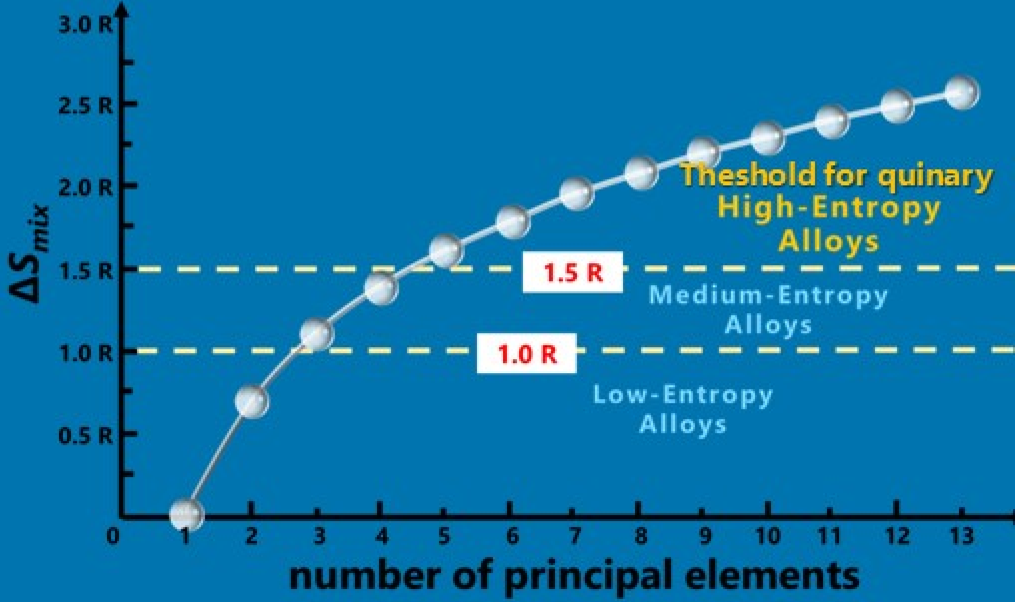
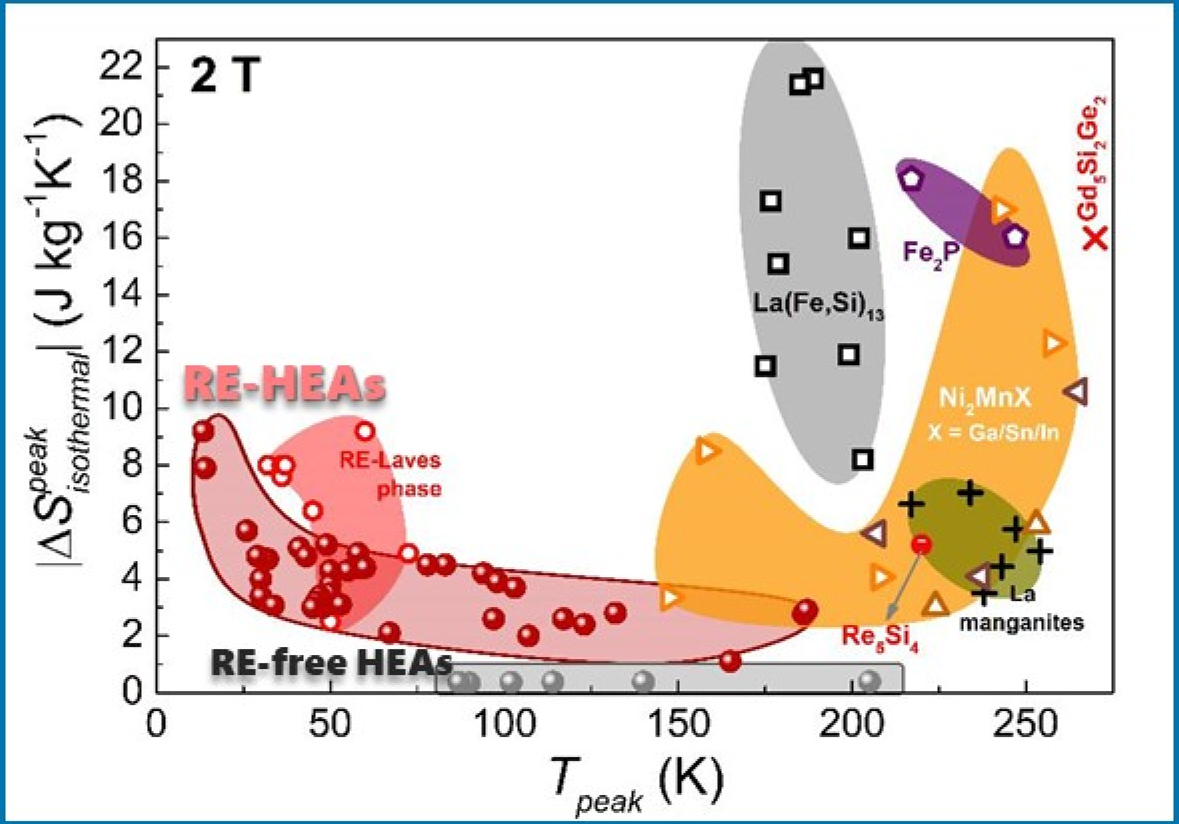


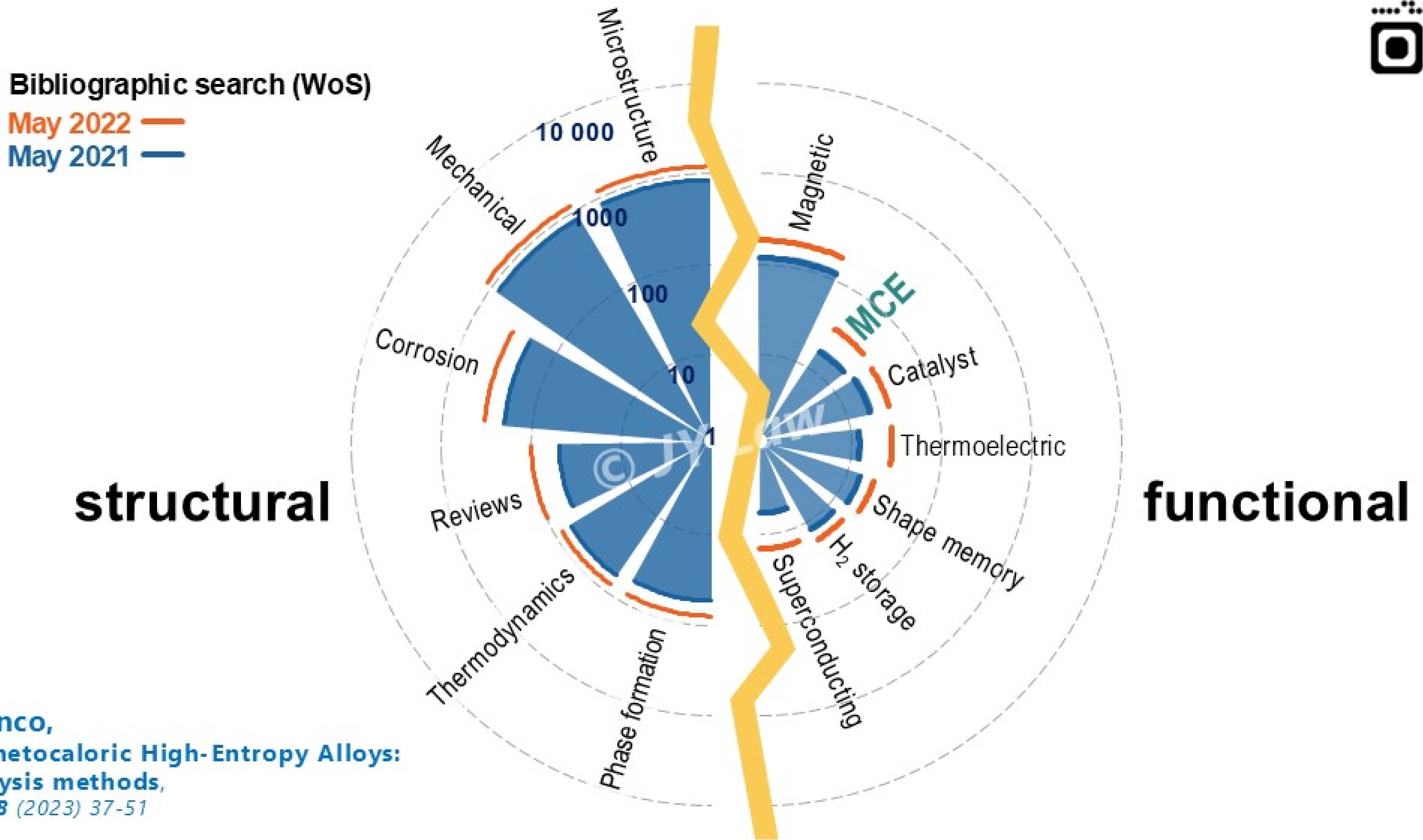
HEAs are far from being competitive

compared to conventional magnetocaloric materials



HEA publications

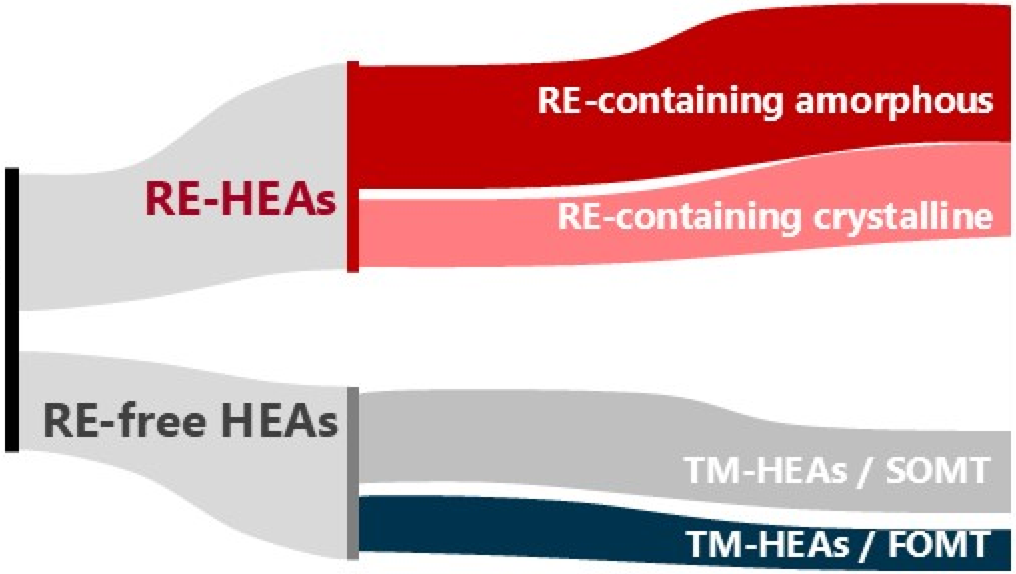
and their different focus



JY Law, V. Franco,
Review on Magnetocaloric High-Entropy Alloys: design and analysis methods,
J Mater Research ***38*** *(2023) 37-51*

Magnetocaloric HEAs

in the literature




APL Materials RESEARCH UPDATE

Pushing the limits of magnetocaloric high-entropy alloys

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Submitted: 29 May 2021 • Accepted: 9 August 2021 •
Published Online: 23 August 2021

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ABSTRACT

High-entropy alloys (HEAs) have become a topic of high research interest due to the excellent mechanical properties that can be found in this new type of materials. However, their functional properties are usually modest when compared to conventional materials. The discovery of high-entropy alloys with an optimal combination of mechanical and functional properties would be a leap forward in the reliability of devices that use them as functional elements. This Research Update focuses on magnetocaloric HEAs, showing that a directed search strategy allows us to improve their performance in a significant way, closing the pre-existing gap between magnetocaloric HEAs and high-performance magnetocaloric materials. Further challenges that remain in this line of research are highlighted.

I. INTRODUCTION

Metallurgy is circa five millennia old, and for the vast majority of its past, it has mainly focused on designing new alloys by adding minor amounts of elements to one or two main constituents. Over time, the possibilities of finding new element combinations that cause a significant enhancement of the materials' properties are getting exhausted, which motivates the search for radically new approaches for alloy development. This change of philosophy arrived with the concept of high-entropy alloys (HEAs), which utilize multiple principal elements (five or more) in relatively high concentrations to form materials with a high entropy of mixing (ΔS_{mix}). HEAs encompass a vast compositional space, which provides a large window of promising opportunities for discovery of new alloys with valuable properties. In general, there are two widely accepted ways of defining HEAs based on the configurational entropy and composition requirements.[1,2]

A. Entropy-based definition

HEAs are defined as alloys with five or more principal elements, exhibiting $\Delta S_{mix} \geq 1.5R$ regardless of being single or multiphase at room temperature, in which ΔS_{mix} is calculated as

$$\Delta S_{mix} = -R \sum x_i \ln x_i, \quad (1)$$

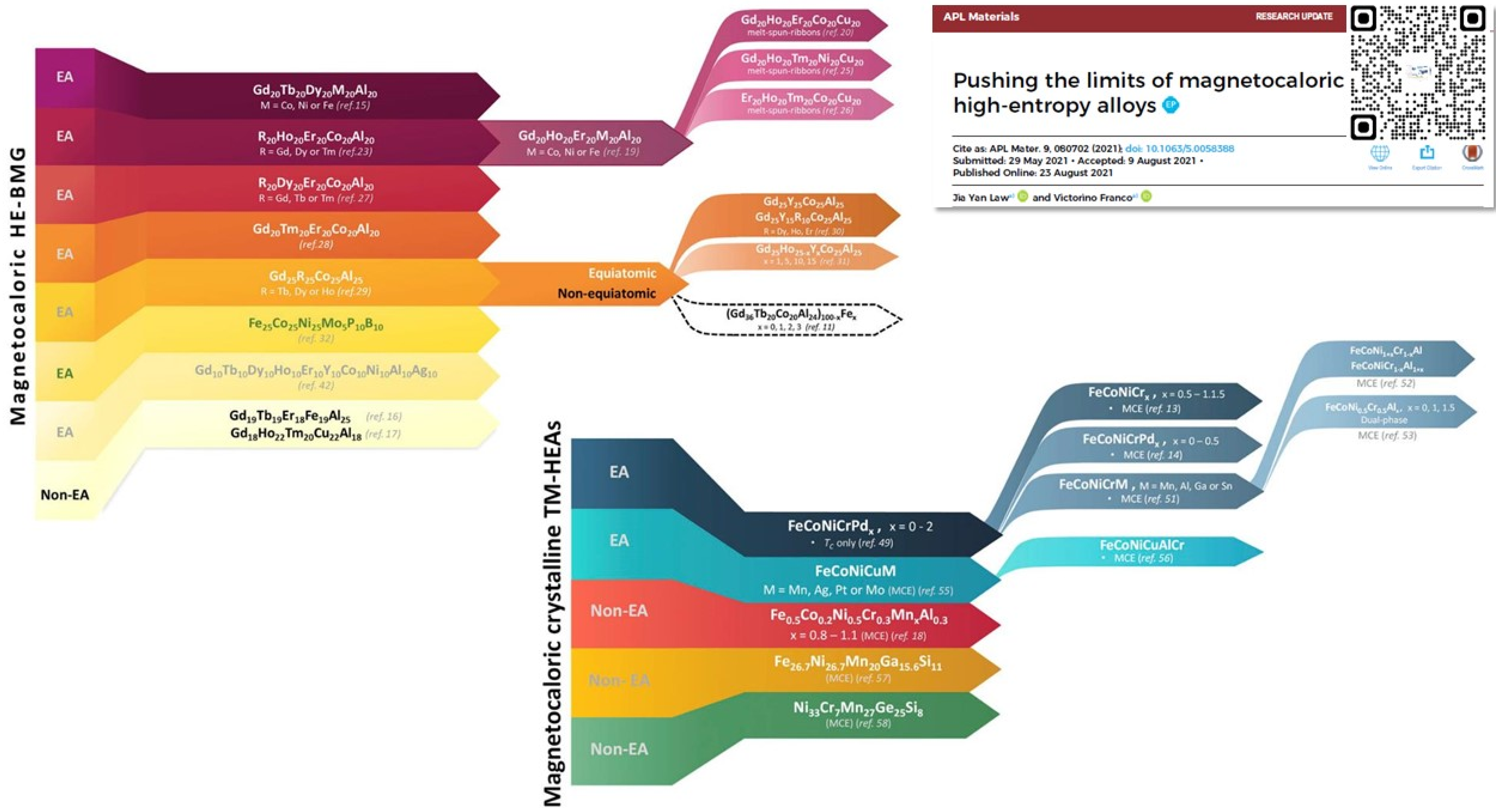
where R is the gas constant and x_i is the mole fraction of the ith element. It is worth noting that the entropy of mixing is usually simplified to exclusively consider the configurational entropy since this term dominates over the other contributions to the total entropy of mixing, which are vibrational, magnetic dipole, and electronic randomness. Figure 1 shows the various classifications of materials based on their configurational entropy values and number of principal elements, where the community preferentially considered that ΔS_{mix} of $1.5R$ is large enough to be used as the boundary between HEAs and medium-entropy alloys.

B. Composition-based definition

An alternate definition, based on the concentration of the different elements, can be summarized as

$$n_{major} \geq 5,\ 5\text{ at. \%} \leq c_i \leq 35\text{ at. \%},$$
$$n_{minor} \geq 0,\ c_j \leq 5\text{ at. \%},$$

where n_{major} and n_{minor} represent the number of major and minor elements, respectively. Minor elements here refer to elemental additives that are usually added to HEA systems for further tuning desired properties. c_i and c_j are the atomic percentage of ith major and jth minor elements, respectively.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RESEARCH UPDATE
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Magnetocaloric HE-BMG
EA
EA
EA
EA
EA
EA
EA
EA
Non-EA
$Gd_{20}Tb_{20}Dy_{20}M_{20}Al_{20}$
M = Co, Ni or Fe (ref.15)
$R_{20}Ho_{20}Er_{20}Co_{20}Al_{20}$
R = Gd, Dy or Tm (ref.23)
$R_{20}Dy_{20}Er_{20}Co_{20}Al_{20}$
R = Gd, Tb or Tm (ref. 27)
$Gd_{20}Tm_{20}Er_{20}Co_{20}Al_{20}$
(ref.28)
$Gd_{25}R_{25}Co_{25}Al_{25}$
R = Tb, Dy or Ho (ref.29)
$Fe_{25}Co_{25}Ni_{25}Mo_{5}P_{10}B_{10}$
(ref. 32)
$Gd_{10}Tb_{10}Dy_{10}Ho_{10}Er_{10}Y_{10}Co_{10}Ni_{10}Al_{10}Ag_{10}$
(ref. 42)
$Gd_{19}Tb_{19}Er_{18}Fe_{19}Al_{25}$ (ref. 16)
$Gd_{18}Ho_{22}Tm_{20}Cu_{22}Al_{18}$ (ref. 17)
$Gd_{20}Ho_{20}Er_{20}M_{20}Al_{20}$
M = Co, Ni or Fe (ref. 19)
$Gd_{20}Ho_{20}Er_{20}Co_{20}Cu_{20}$
melt-spun-ribbons (ref. 20)
$Gd_{20}Ho_{20}Tm_{20}Ni_{20}Cu_{20}$
melt-spun-ribbons (ref. 25)
$Er_{20}Ho_{20}Tm_{20}Co_{20}Cu_{20}$
melt-spun-ribbons (ref. 26)
Equiatomic
Non-equiatomic
$Gd_{25}Y_{25}Co_{25}Al_{25}$
$Gd_{25}Y_{15}R_{10}Co_{25}Al_{25}$
R = Dy, Ho, Er (ref. 30)
$Gd_{25}Ho_{25-x}Y_{x}Co_{25}Al_{25}$
x = 1, 5, 10, 15 (ref. 31)
$(Gd_{36}Tb_{20}Co_{20}Al_{24})_{100-x}Fe_{x}$
x = 0, 1, 2, 3 (ref. 11)
Magnetocaloric crystalline TM-HEAs
EA
EA
Non-EA
Non- EA
Non-EA
$FeCoNiCrPd_{x}$, x = 0 - 2
• T_C only (ref. 49)
FeCoNiCuM
M = Mn, Ag, Pt or Mo (MCE) (ref. 55)
$Fe_{0.5}Co_{0.2}Ni_{0.5}Cr_{0.3}Mn_{x}Al_{0.3}$
x = 0.8 – 1.1 (MCE) (ref. 18)
$Fe_{26.7}Ni_{26.7}Mn_{20}Ga_{15.6}Si_{11}$
(MCE) (ref. 57)
$Ni_{33}Cr_{7}Mn_{27}Ge_{25}Si_{8}$
(MCE) (ref. 58)
$FeCoNiCr_{x}$, x = 0.5 – 1.1.5
• MCE (ref. 13)
$FeCoNiCrPd_{x}$, x = 0 – 0.5
• MCE (ref. 14)
FeCoNiCrM, M = Mn, Al, Ga or Sn
• MCE (ref. 51)
FeCoNiCuAlCr
• MCE (ref. 56)
$FeCoNi_{1+x}Cr_{1-x}Al$
$FeCoNiCr_{1-x}Al_{1+x}$
MCE (ref. 52)
$FeCoNi_{0.5}Cr_{0.5}Al_{x}$, x = 0, 1, 1.5
Dual-phase
MCE (ref. 53)

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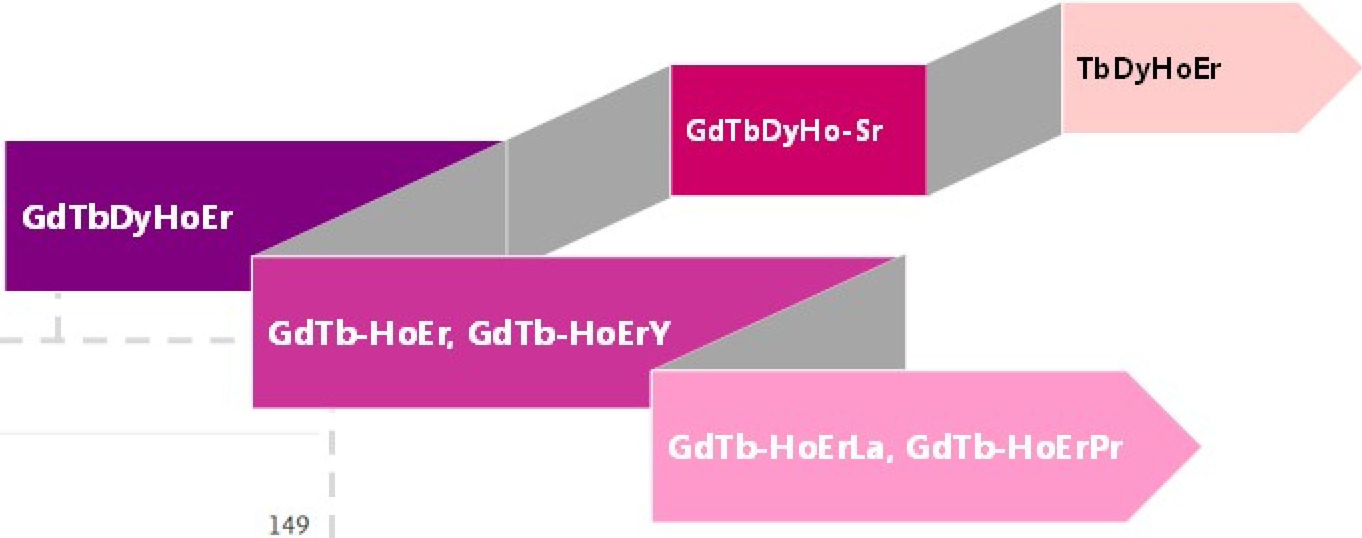

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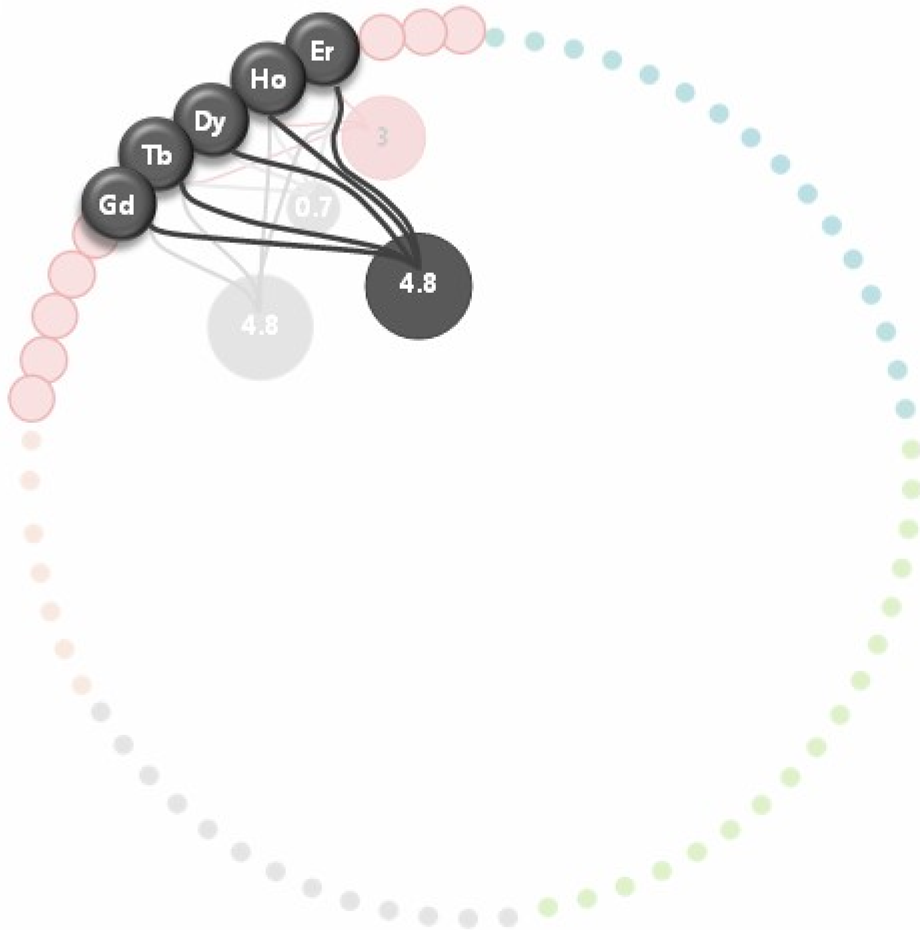
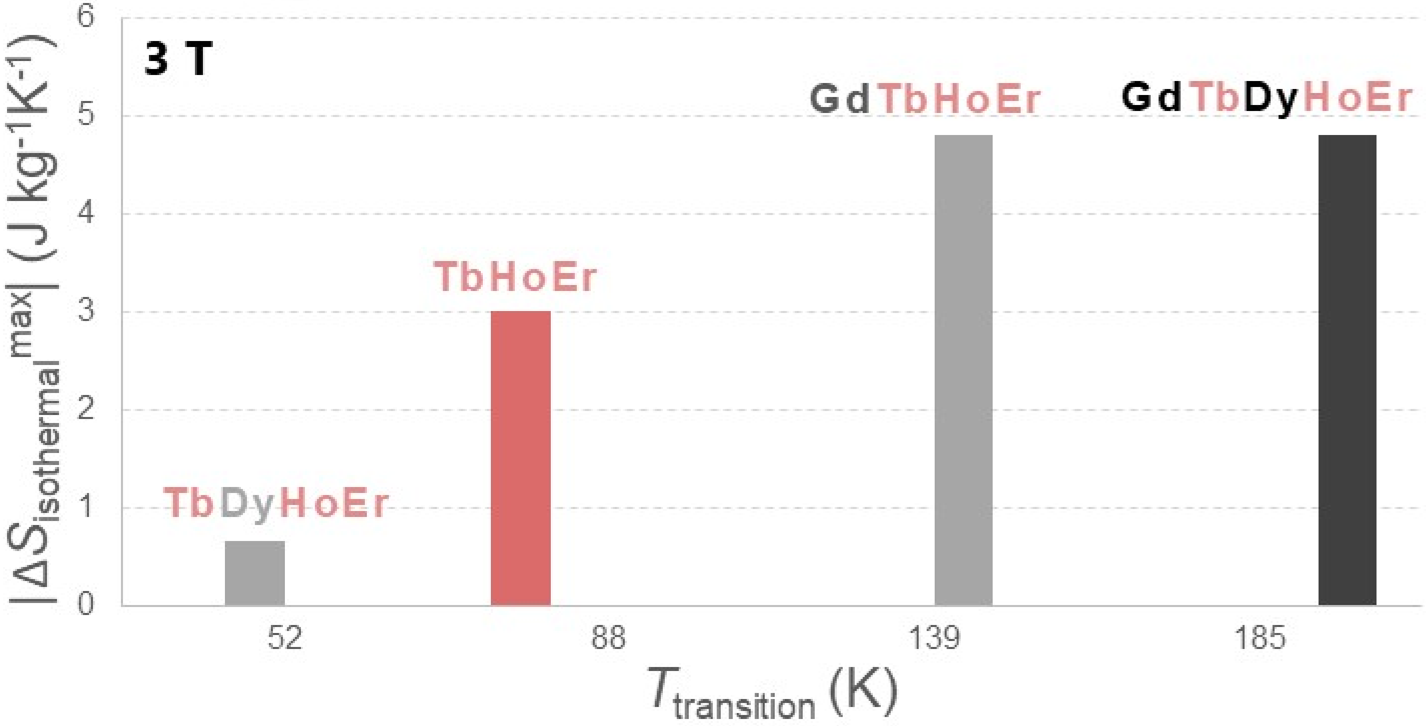
Historical data only

Search strategy

through the large HEA space



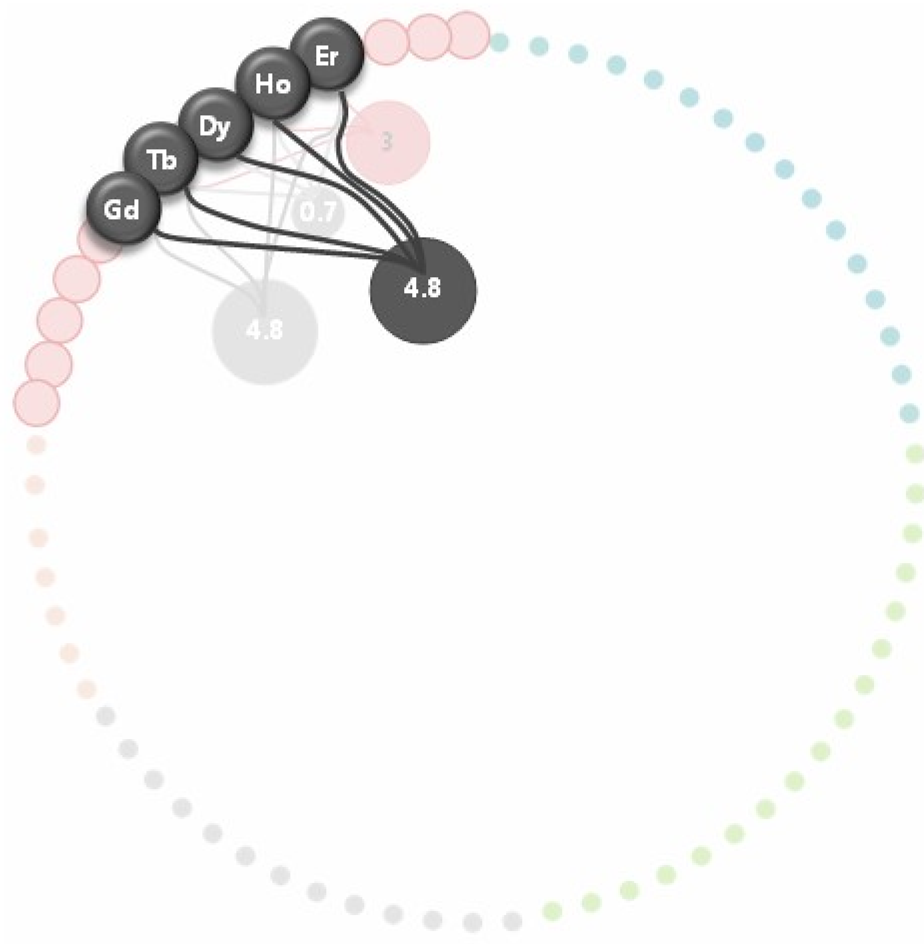
increasing number of principal elements
Rare-earth Elements



Data collected from
Y. Yuan et al., Acta Mater 125 (2017) 481

Search strategy

through the large HEA space



increasing number of principal elements

Rare-earth Elements

Data collected from
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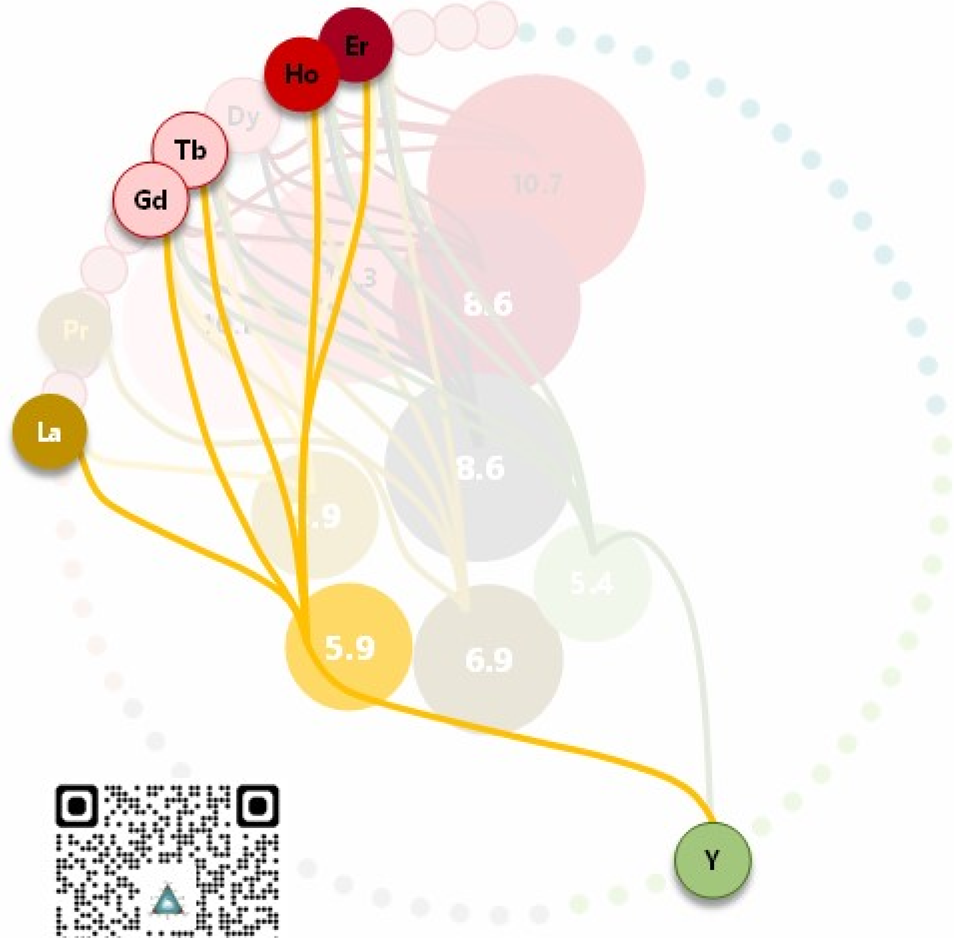
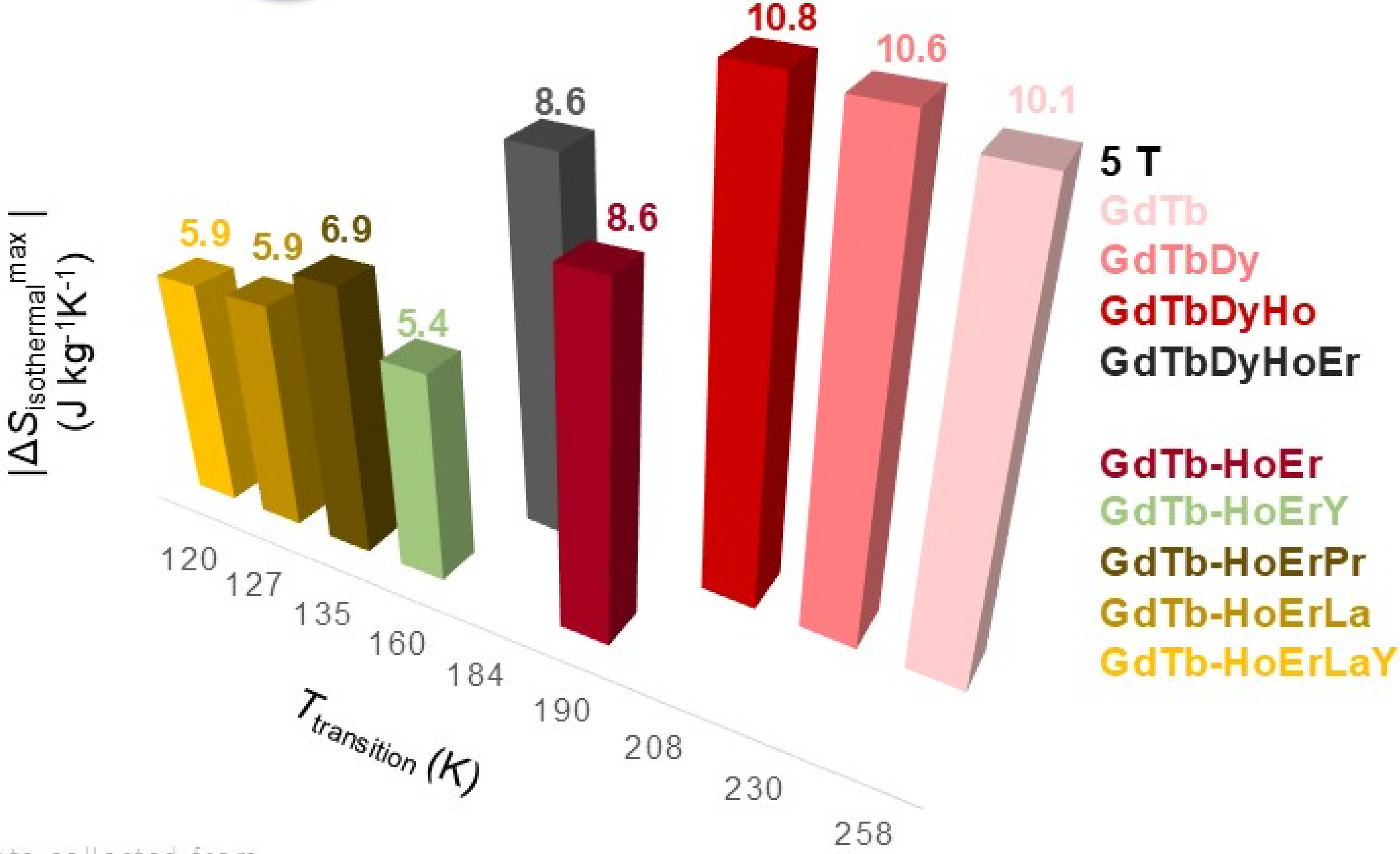
Search strategy

through the large HEA space



increasing number of principal elements



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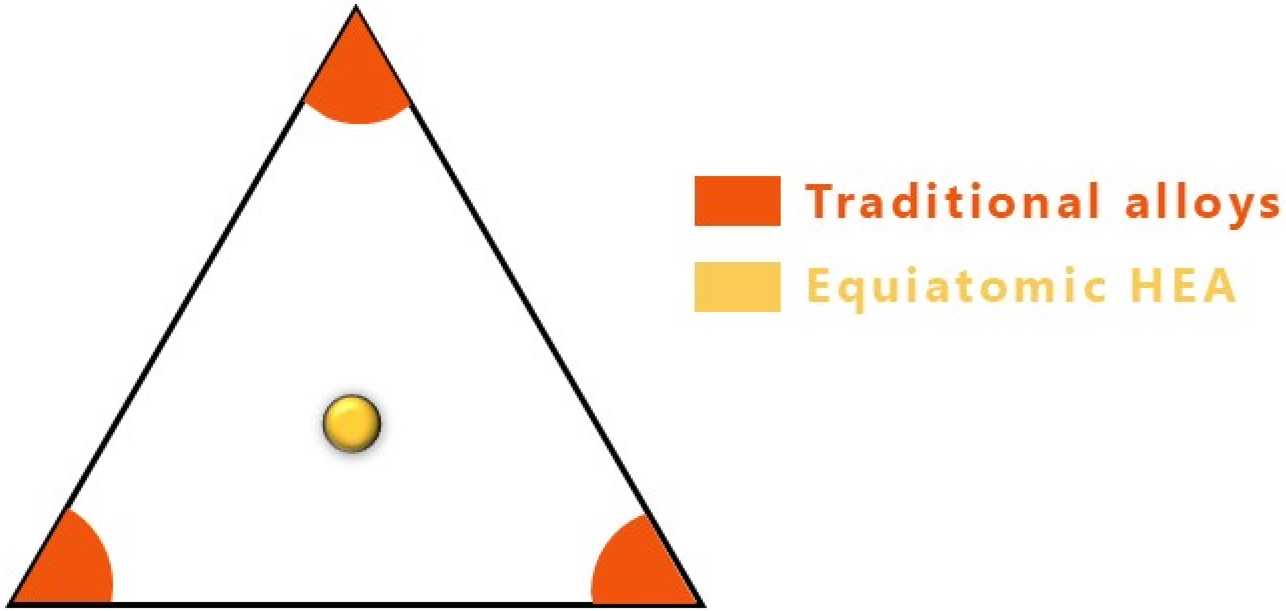
Search strategy

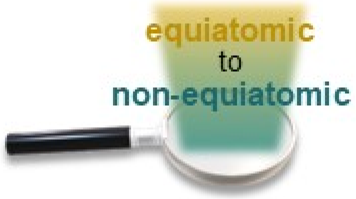
through the large HEA space

increasing number of principal elements

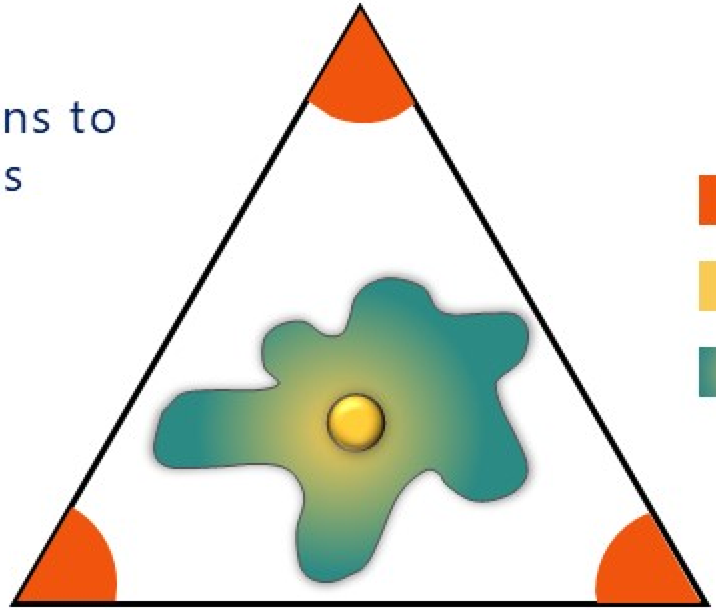


Search strategy

through the large HEA space



extending from
equiatomic compositions to non-equiatomic regions



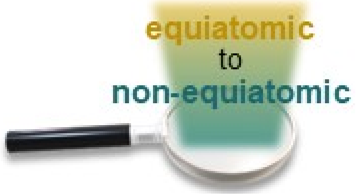
Traditional alloys

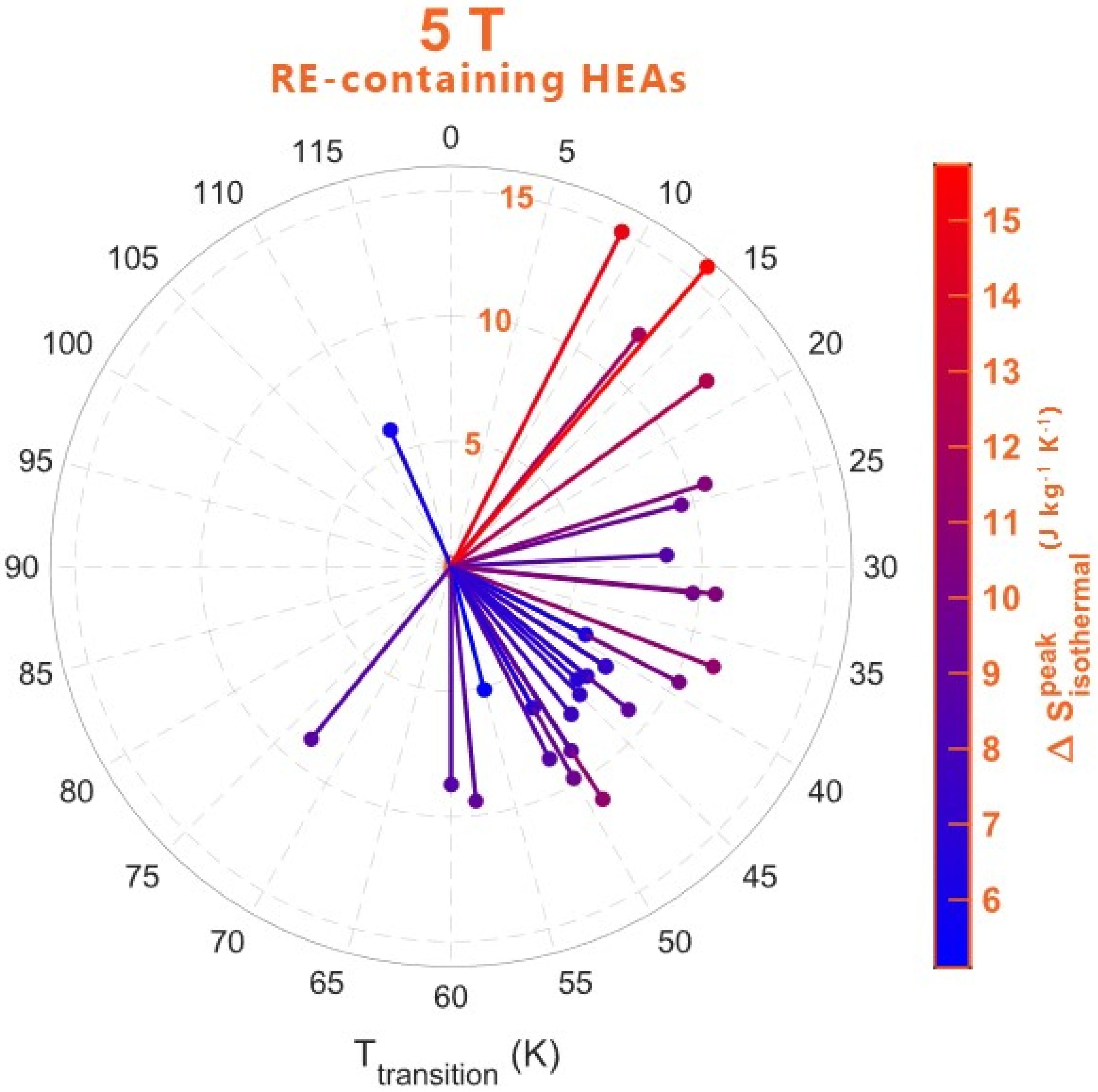
Equiatomic HEA

Non-equiatomic HEA

Search strategy

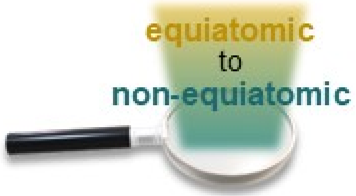
through the large HEA space

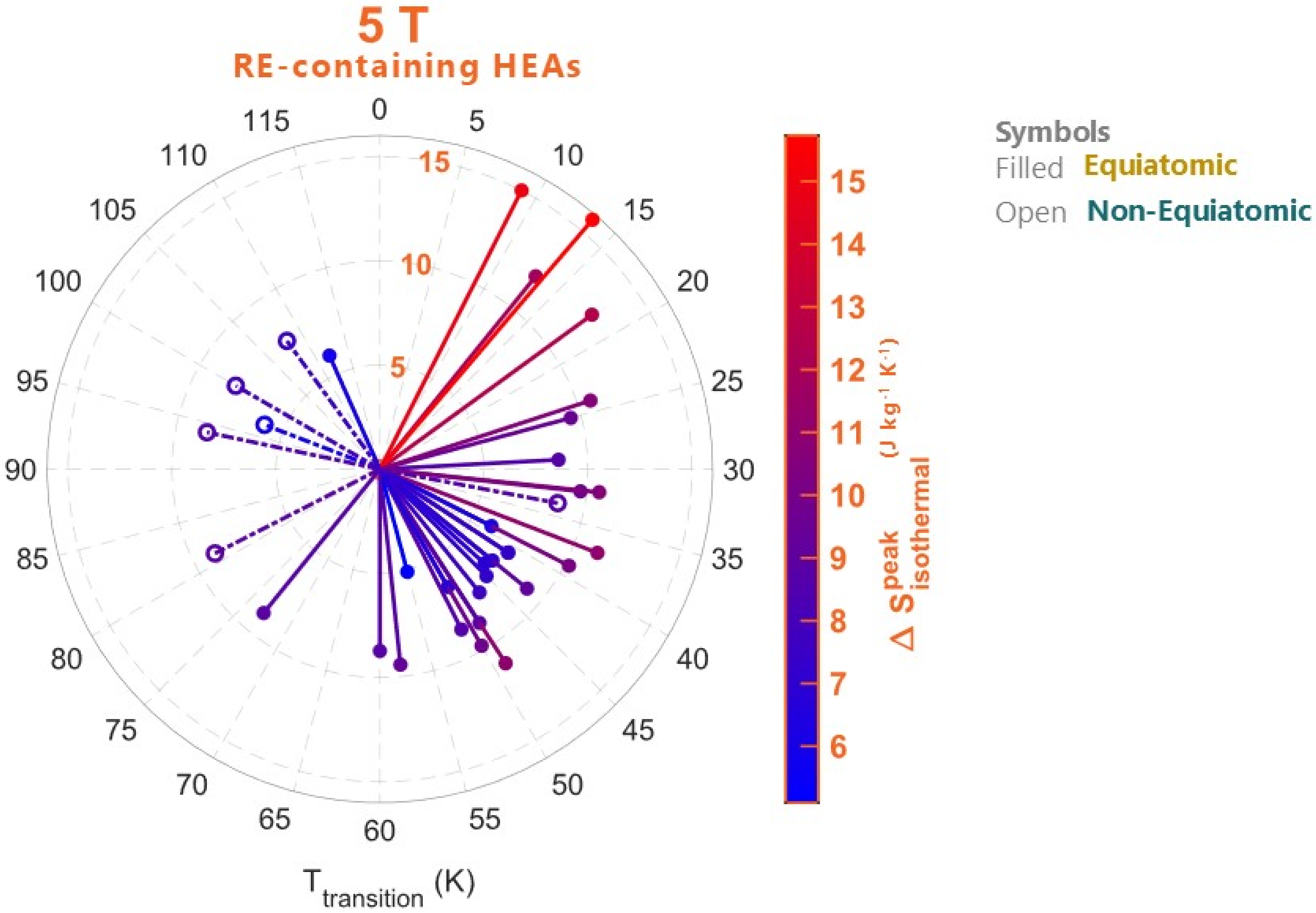




Search strategy

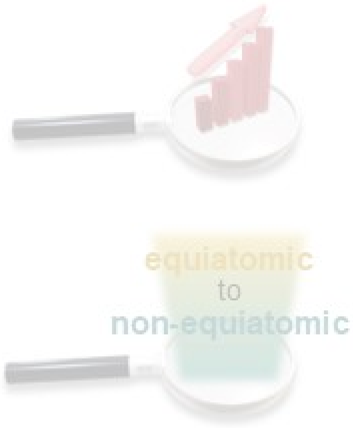
through the large HEA space

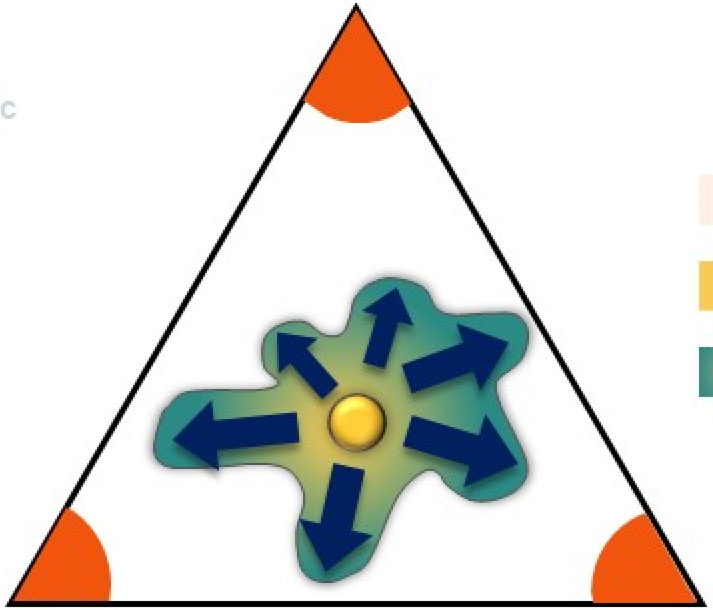




Search strategy

through the large HEA space





Traditional alloys

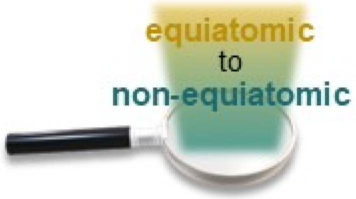
Equiatomic HEA

Non-equiatomic HEA

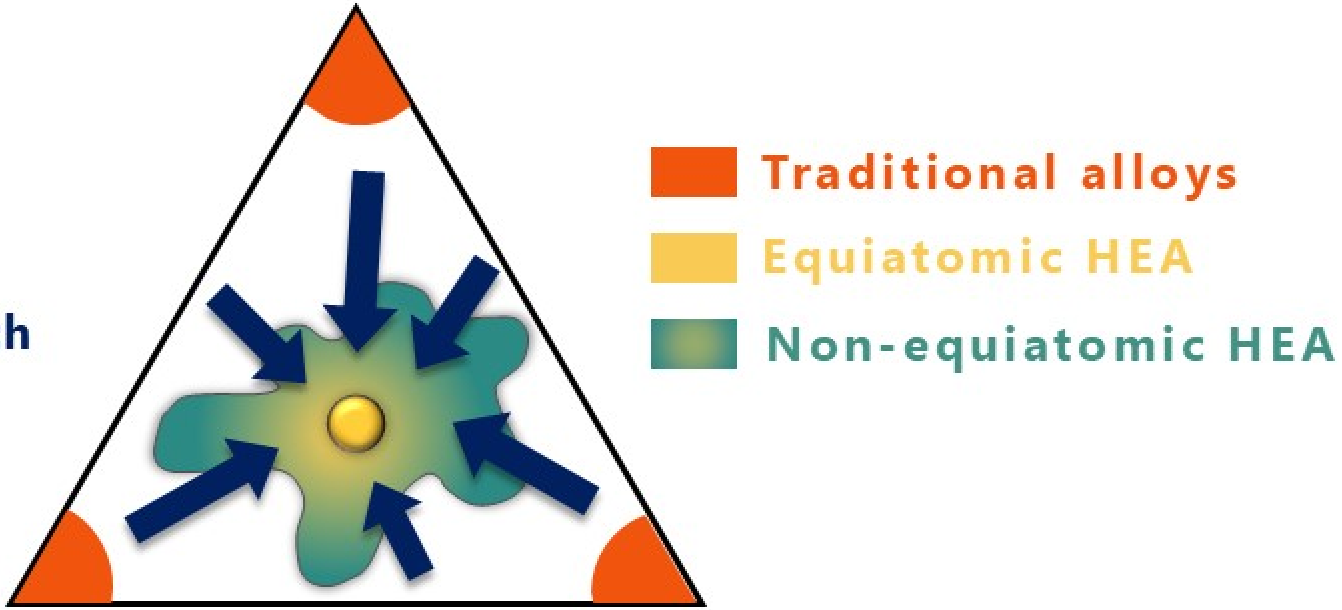
SOPT

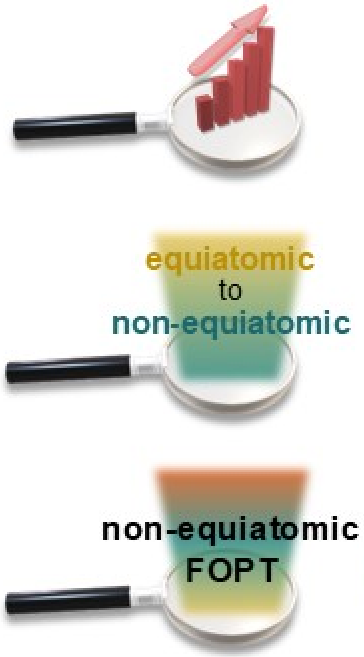
Search strategy

through the large HEA space

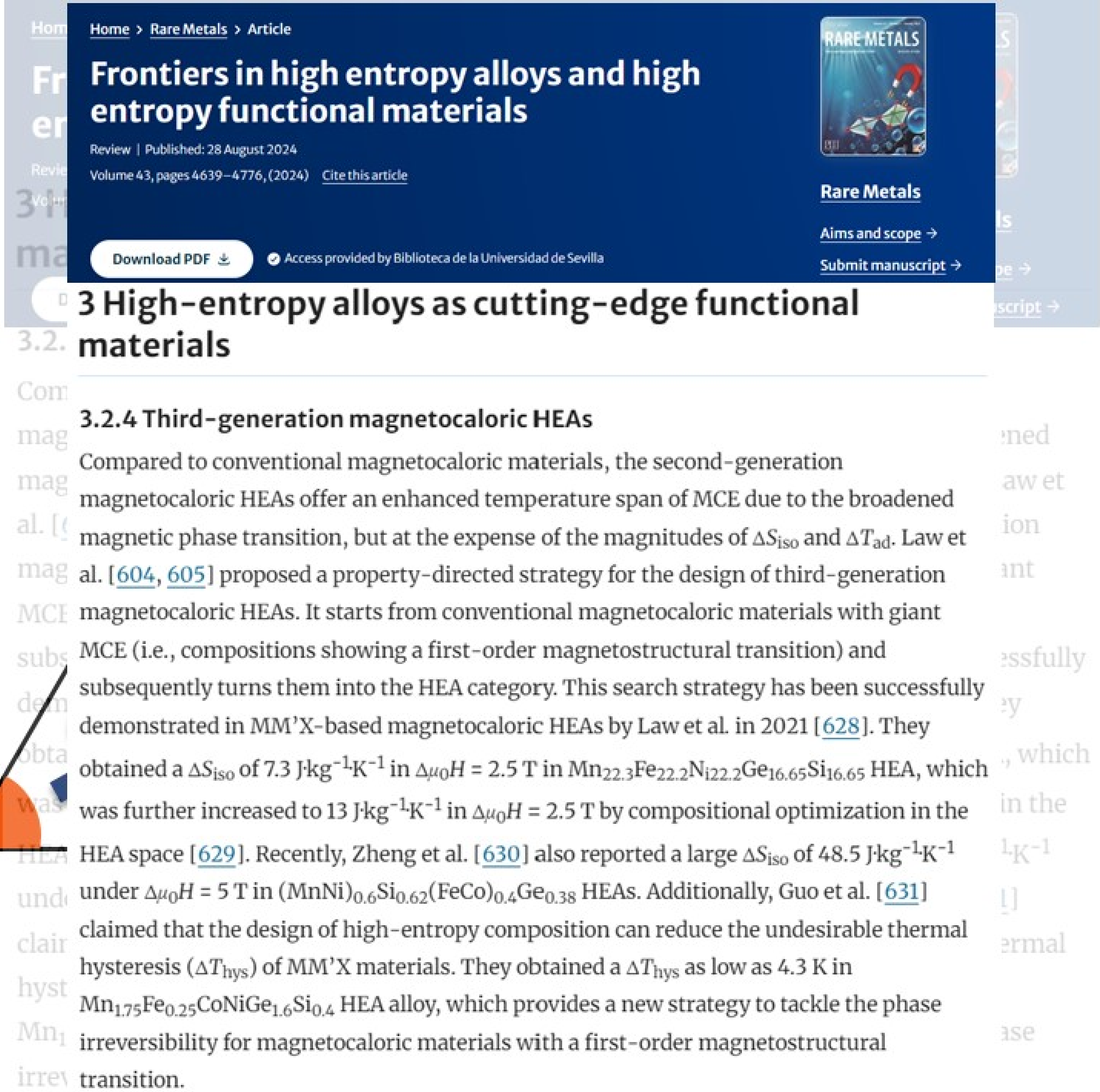


targeted property search





targeted property search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Frontiers in high entropy alloys and high entropy functional materials

Review | Published: 28 August 2024

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Rare Metals

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3 High-entropy alloys as cutting-edge functional materials

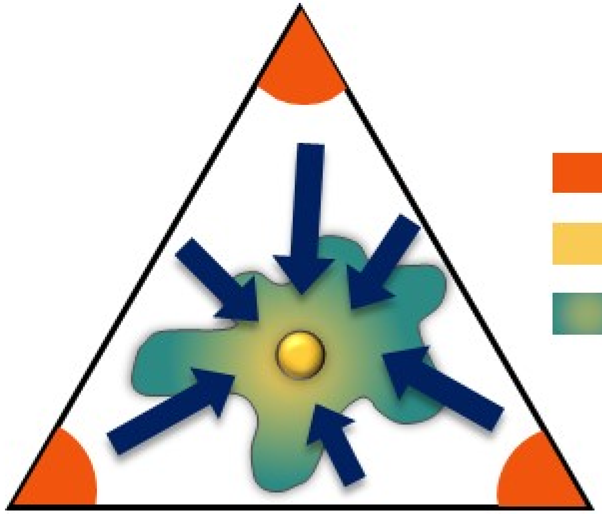
3.2.4 Third-generation magnetocaloric HEAs

Compared to conventional magnetocaloric materials, the second-generation magnetocaloric HEAs offer an enhanced temperature span of MCE due to the broadened magnetic phase transition, but at the expense of the magnitudes of ΔS_{iso} and ΔT_{ad}. Law et al. [604, 605] proposed a property-directed strategy for the design of third-generation magnetocaloric HEAs. It starts from conventional magnetocaloric materials with giant MCE (i.e., compositions showing a first-order magnetostructural transition) and subsequently turns them into the HEA category. This search strategy has been successfully demonstrated in MM'X-based magnetocaloric HEAs by Law et al. in 2021 [628]. They obtained a ΔS_{iso} of 7.3 J·kg^{-1}K^{-1} in $\Delta\mu_0 H$ = 2.5 T in $Mn_{22.3}Fe_{22.2}Ni_{22.2}Ge_{16.65}Si_{16.65}$ HEA, which was further increased to 13 J·kg^{-1}K^{-1} in $\Delta\mu_0 H$ = 2.5 T by compositional optimization in the HEA space [629]. Recently, Zheng et al. [630] also reported a large ΔS_{iso} of 48.5 J·kg^{-1}K^{-1} under $\Delta\mu_0 H$ = 5 T in $(MnNi)_{0.6}Si_{0.62}(FeCo)_{0.4}Ge_{0.38}$ HEAs. Additionally, Guo et al. [631] claimed that the design of high-entropy composition can reduce the undesirable thermal hysteresis (ΔT_{hys}) of MM'X materials. They obtained a ΔT_{hys} as low as 4.3 K in $Mn_{1.75}Fe_{0.25}CoNiGe_{1.6}Si_{0.4}$ HEA alloy, which provides a new strategy to tackle the phase irreversibility for magnetocaloric materials with a first-order magnetostructural transition.

Search strategy

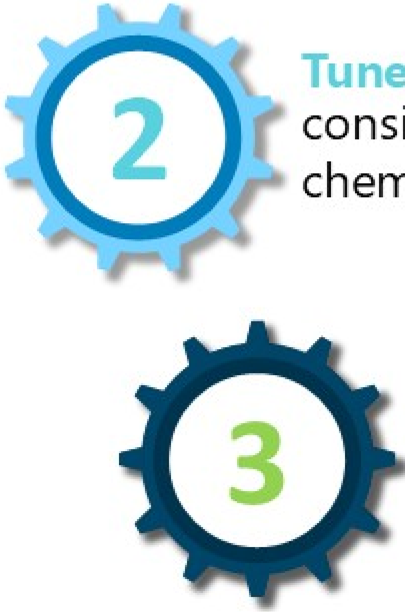
through the large HEA space

targeted property search



- Traditional alloys
- Equiatomic HEA
- Non-equiatomic HEA

Starting composition
selection based on known alloy with desired behavior

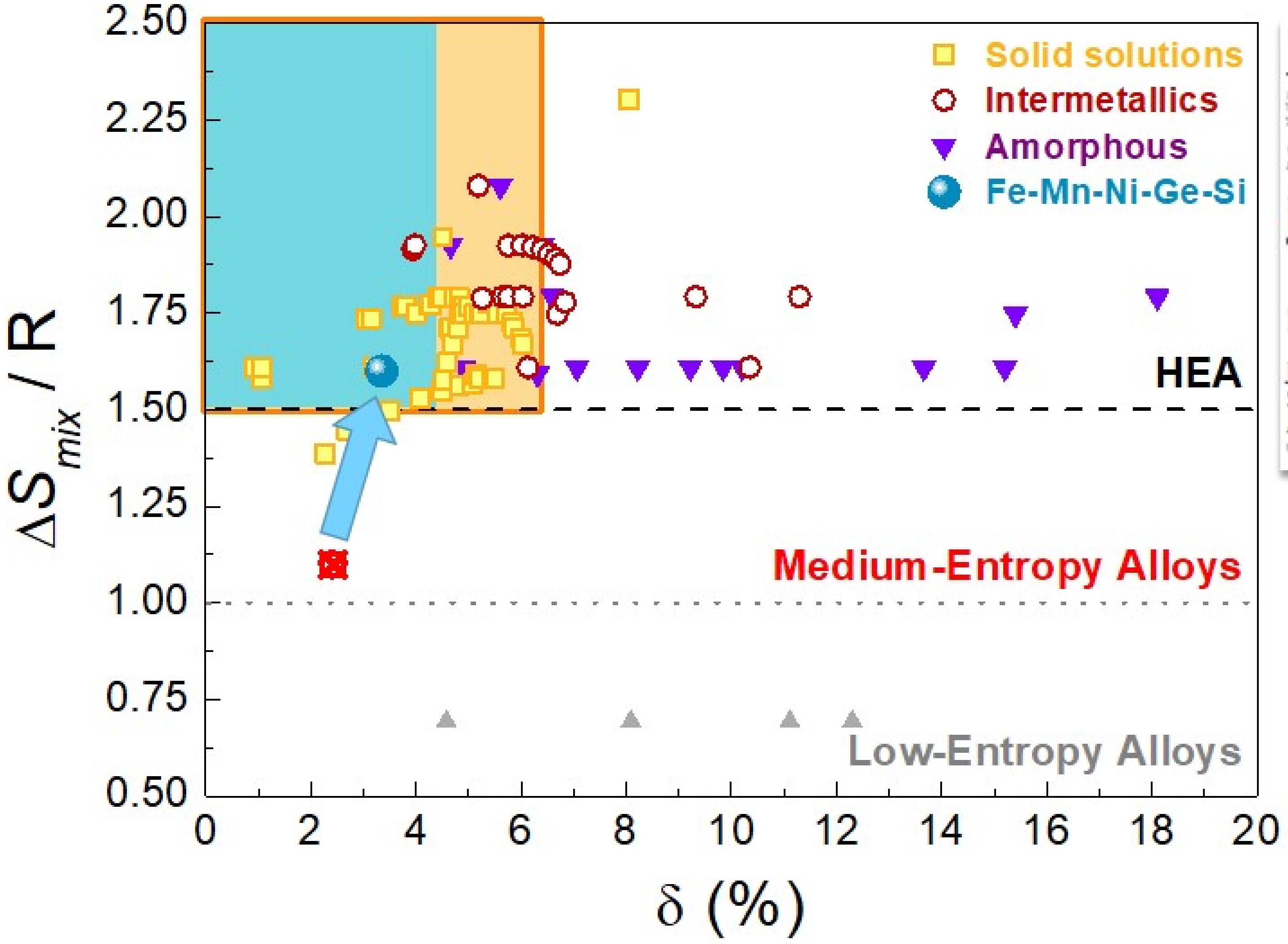


Tune towards HEA region
considering elements with chemical compatibility

Maintain the stoichiometry

Search strategy

in the vast HEA space



Journal of Alloys and Compounds 855 (2021) 157424

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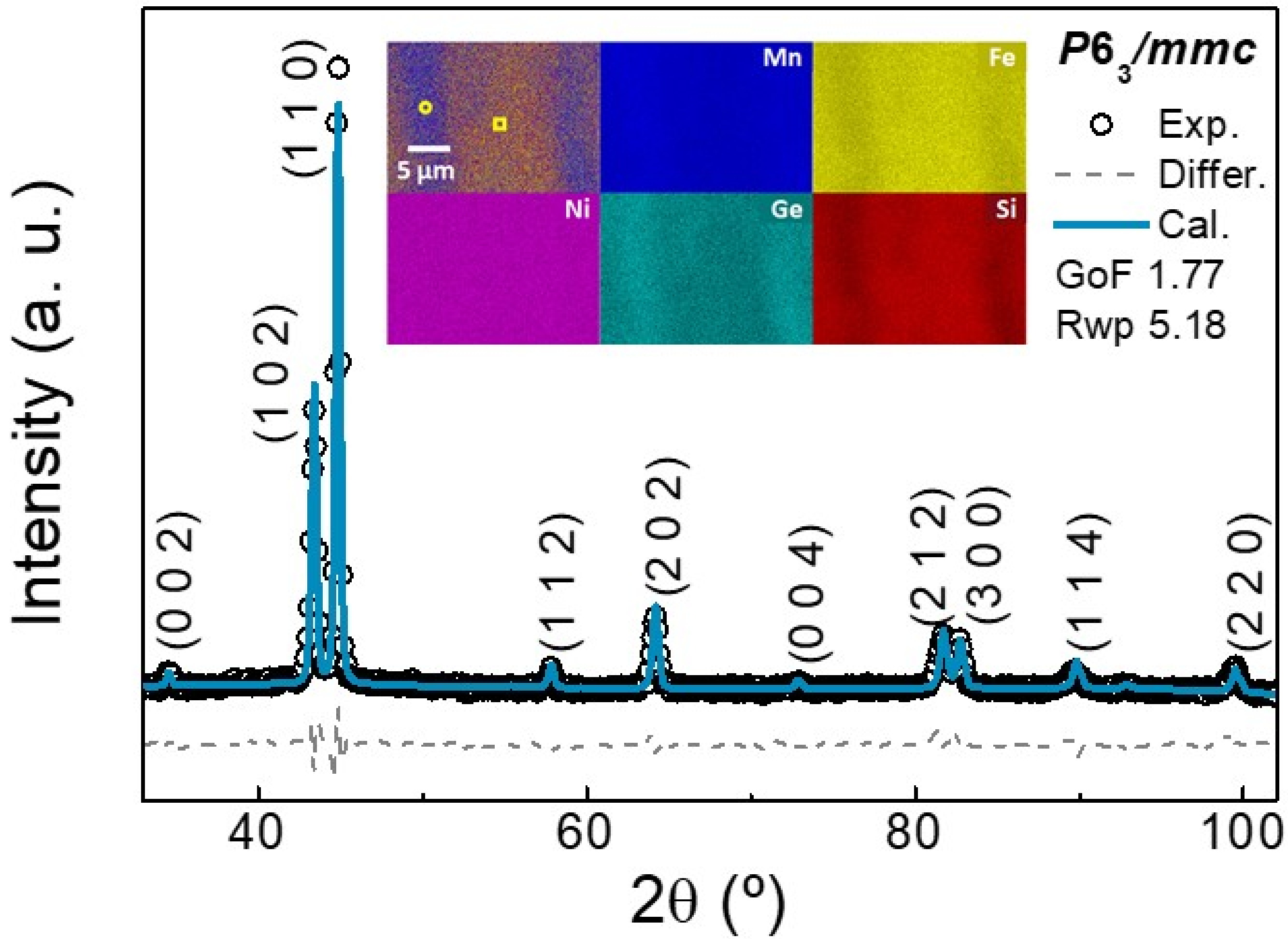
MnFeNiGeSi high-entropy alloy with large magnetocaloric effect

Jia Yan Law [a, **], Luis M. Moreno-Ramírez [a], Álvaro Díaz-García [a], Andrés Martín-Cid [b], Shintaro Kobayashi [b], Shogo Kawaguchi [b], Tetsuya Nakamura [b], Victorino Franco [a, *]

[a] Department of Condensed Matter Physics, ICMS-CSIC, Universidad de Sevilla, P.O. Box 1065, 41080, Sevilla, Spain
[b] SPring-8, Japan Synchrotron Radiation Research Institute, 1-1-1 Kouto, Sayo, Japan

Fe-Mn-Ni-Ge-Si

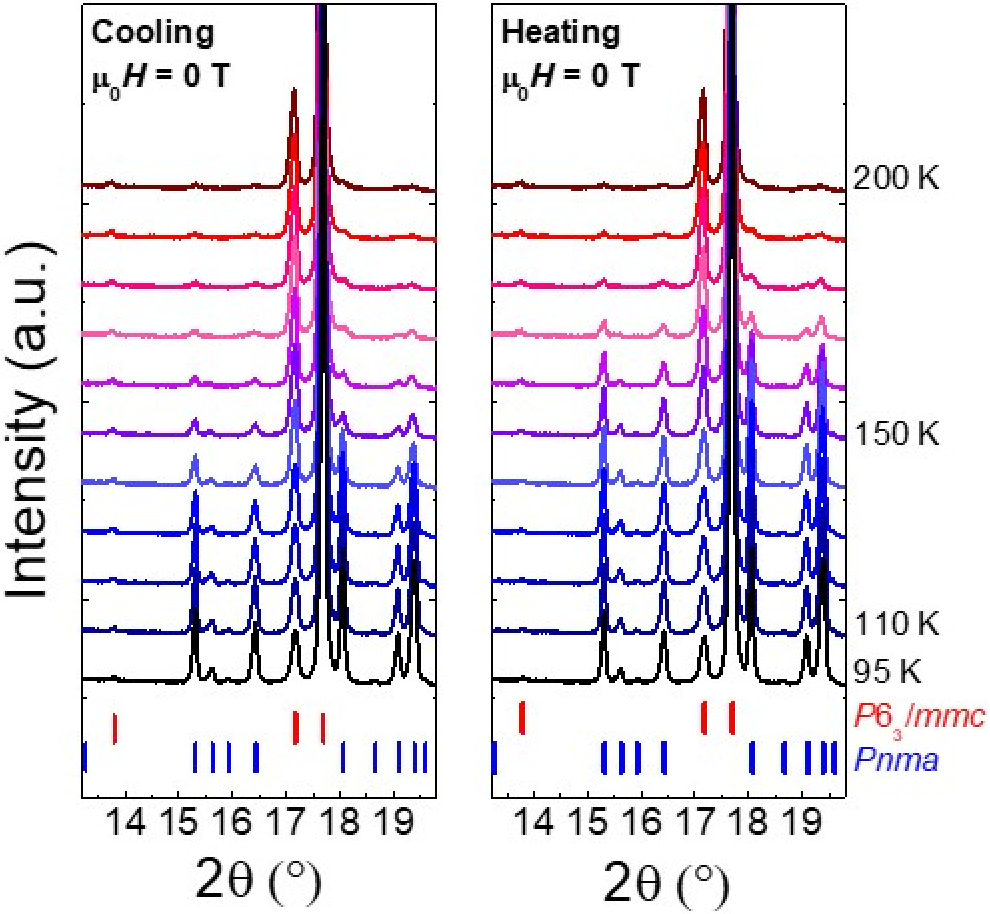
At room temperature

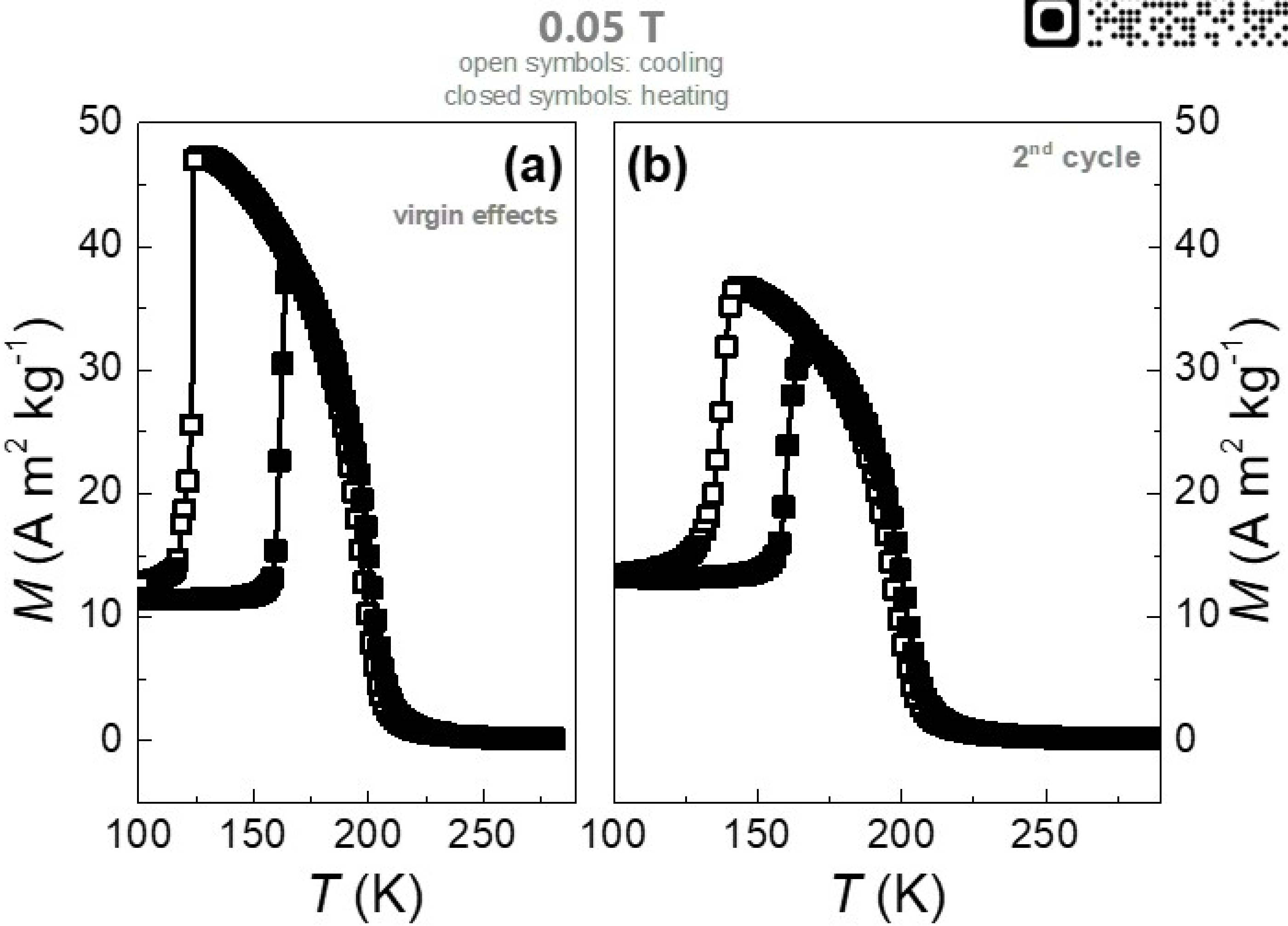


Fe-Mn-Ni-Ge-Si

Upon heating and cooling

JY Law, L.M. Moreno-Ramírez, Á. Díaz-García, A. Martín-Cid, S. Kobayashi, S. Kawaguchi, T. Nakamura and V. Franco, **J Alloys Compds 855 (2021) 157424**

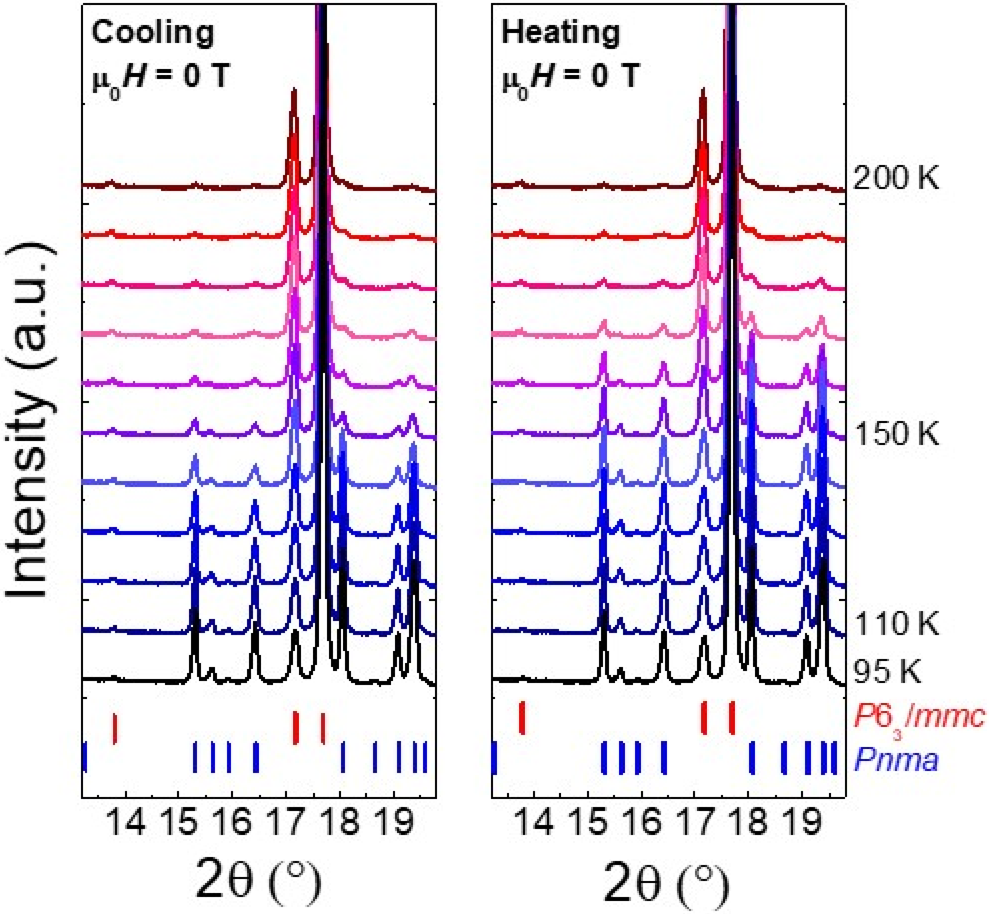


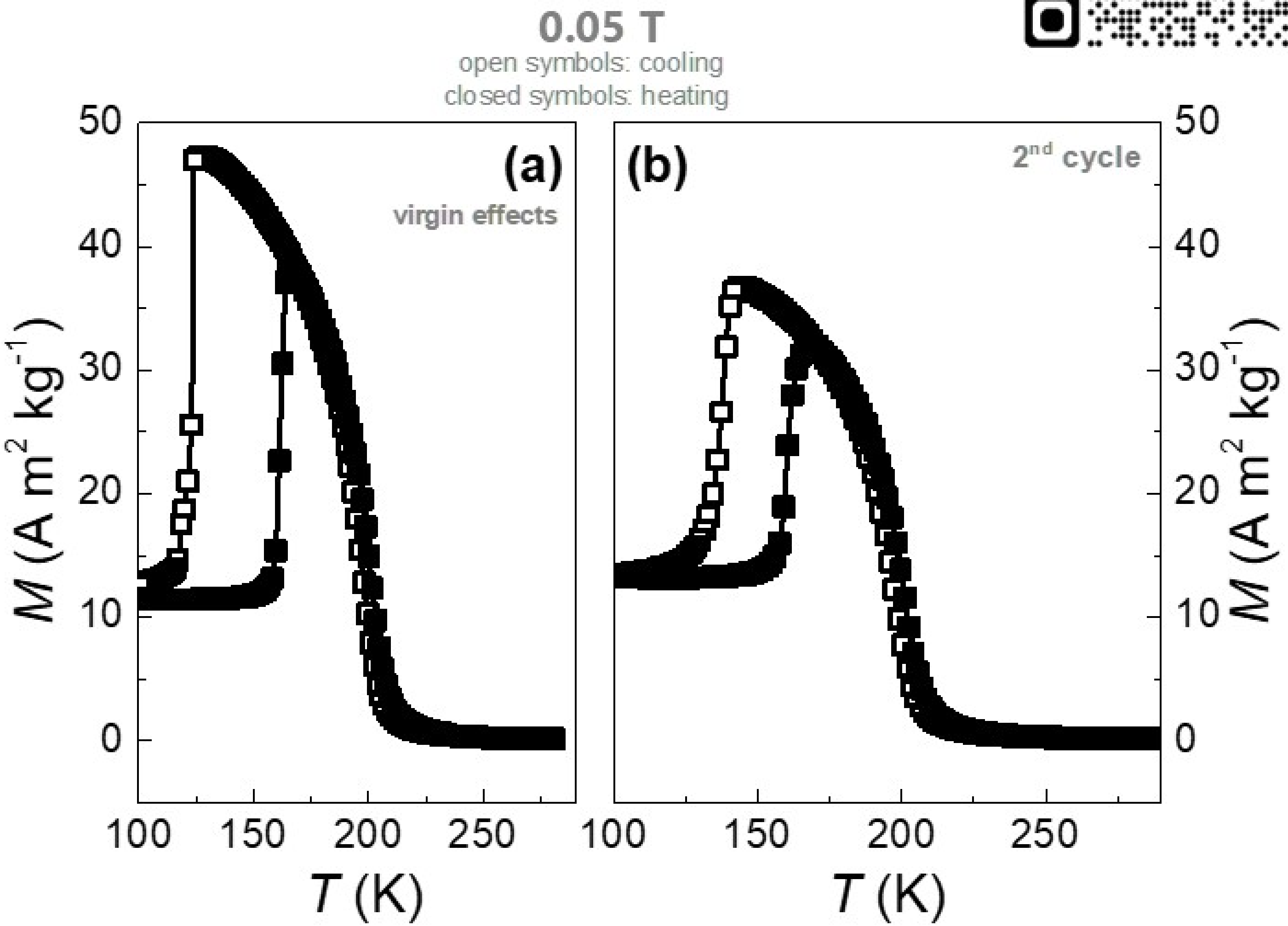


Phase transition temperatures in good agreement

magnetostructural

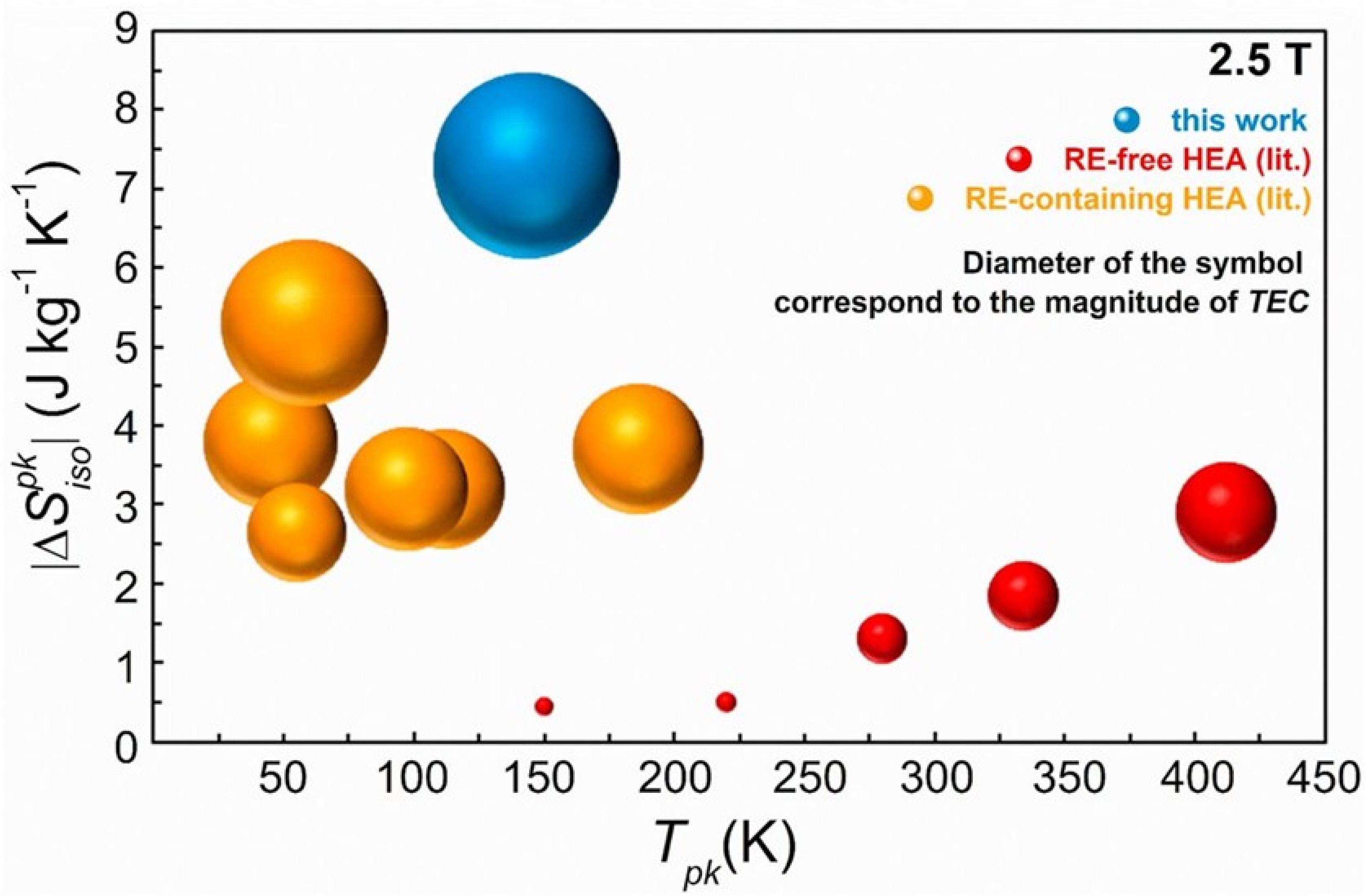
JY Law, L.M. Moreno-Ramírez, Á. Díaz-García, A. Martín-Cid, S. Kobayashi, S. Kawaguchi, T. Nakamura and V. Franco, **J Alloys Compds 855 (2021) 157424**





Fe-Mn-Ni-Ge-Si

MCE



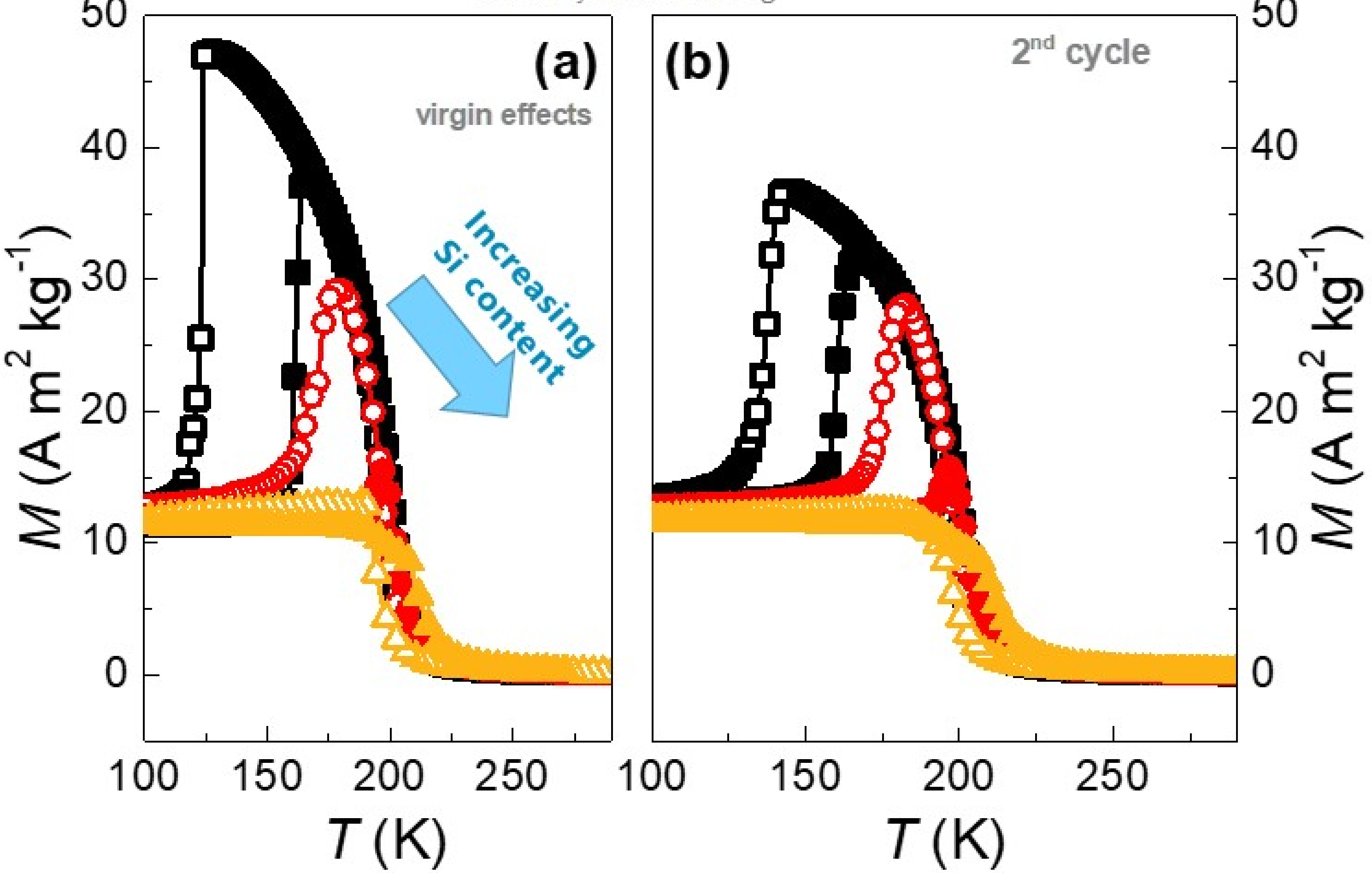
Could that go further?

series (varying Si content)

0.05 T

open symbols: cooling

closed symbols: heating



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Full length article

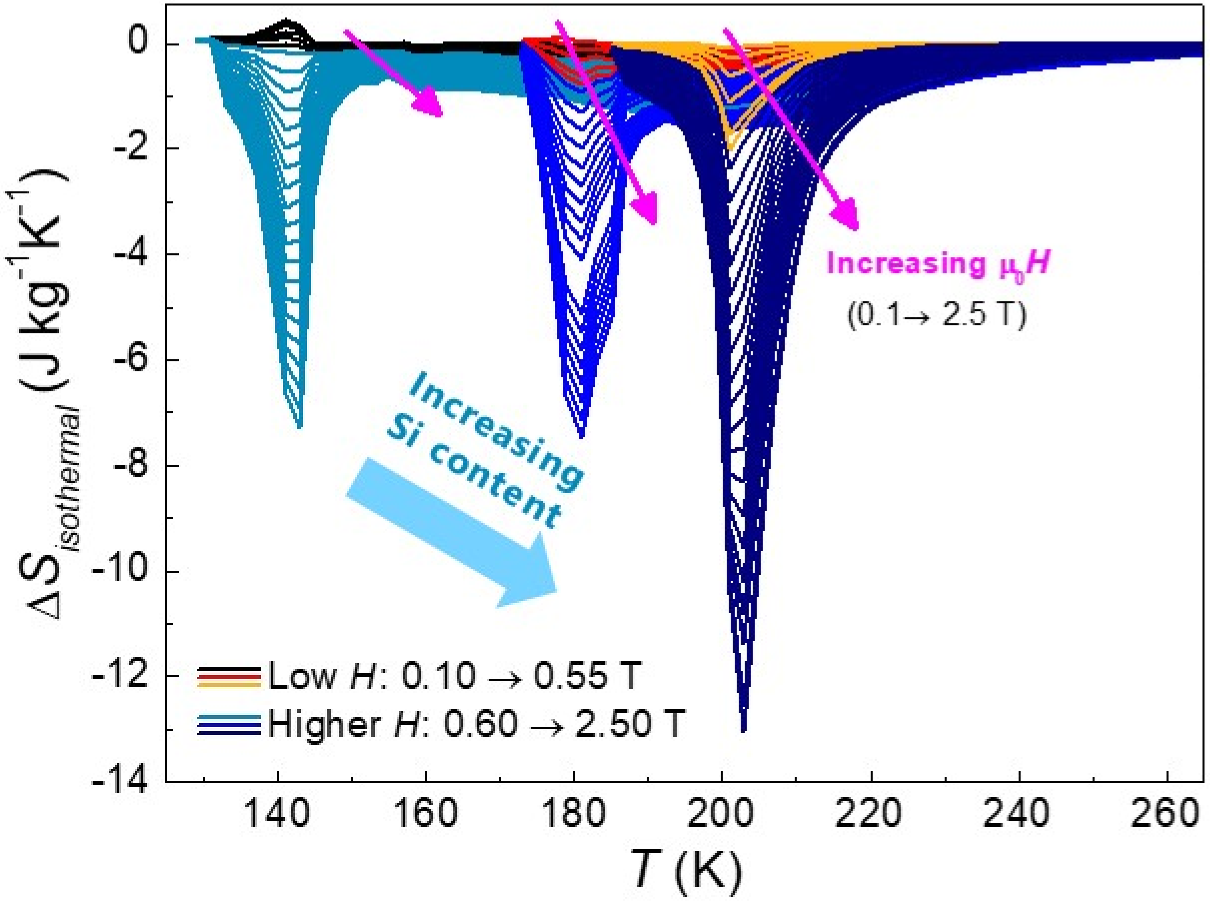
Increased magnetocaloric response of FeMnNiGeSi high-entropy alloys

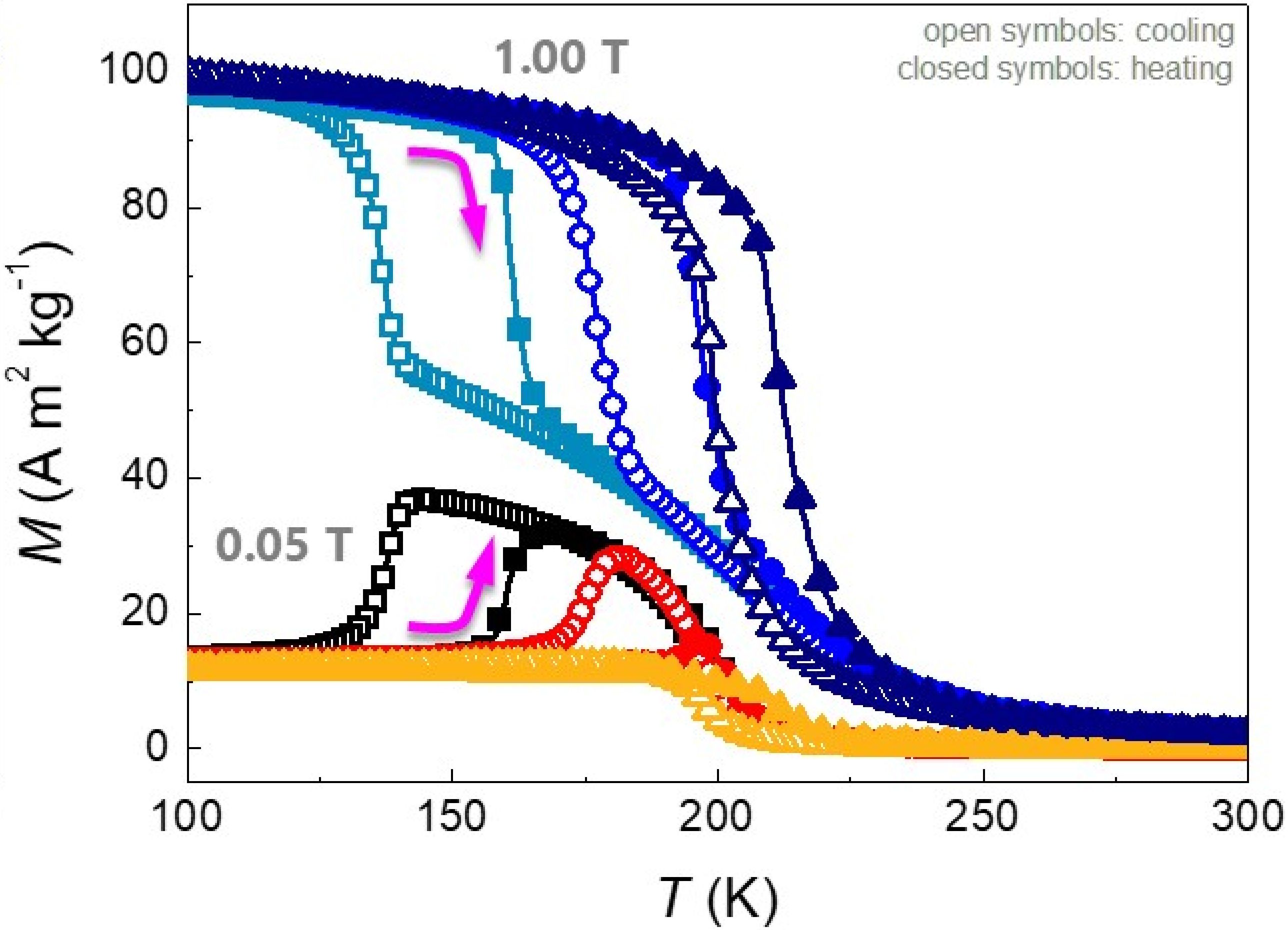
Jia Yan Law*, Álvaro Díaz-García, Luis M. Moreno-Ramírez, Victorino Franco*

Departamento de Física de la Materia Condensada, ICMS-CSIC, Universidad de Sevilla, P.O. Box 1065, 41080-Sevilla, Spain

Fe-Mn-Ni-Ge-Si series

High and low fields





Fe-Mn-Ni-Ge-Si

their order of phase transitions?

to explore using their exponent *n*

nature COMMUNICATIONS

Nat Commun 9 (2018) 2680

ARTICLE

DOI: 10.1038/s41467-018-05111-w OPEN

A quantitative criterion for determining the order of magnetic phase transitions using the magnetocaloric effect

Jia Yan Law[1], Victorino Franco[1], Luis Miguel Moreno-Ramírez[1], Alejandro Conde[1], Dmitriy Y. Karpenkov[2], Iliya Radulov[2], Konstantin P. Skokov[2] & Oliver Gutfleisch[2]

order of phase transition

can be determined by exponent *n*


nature COMMUNICATIONS

ARTICLE

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A quantitative criterion for determining the order of magnetic phase transitions using the magnetocaloric effect

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$$\Delta S_{isothermal} \propto H^{\,n}$$

where n depends on temperature and field

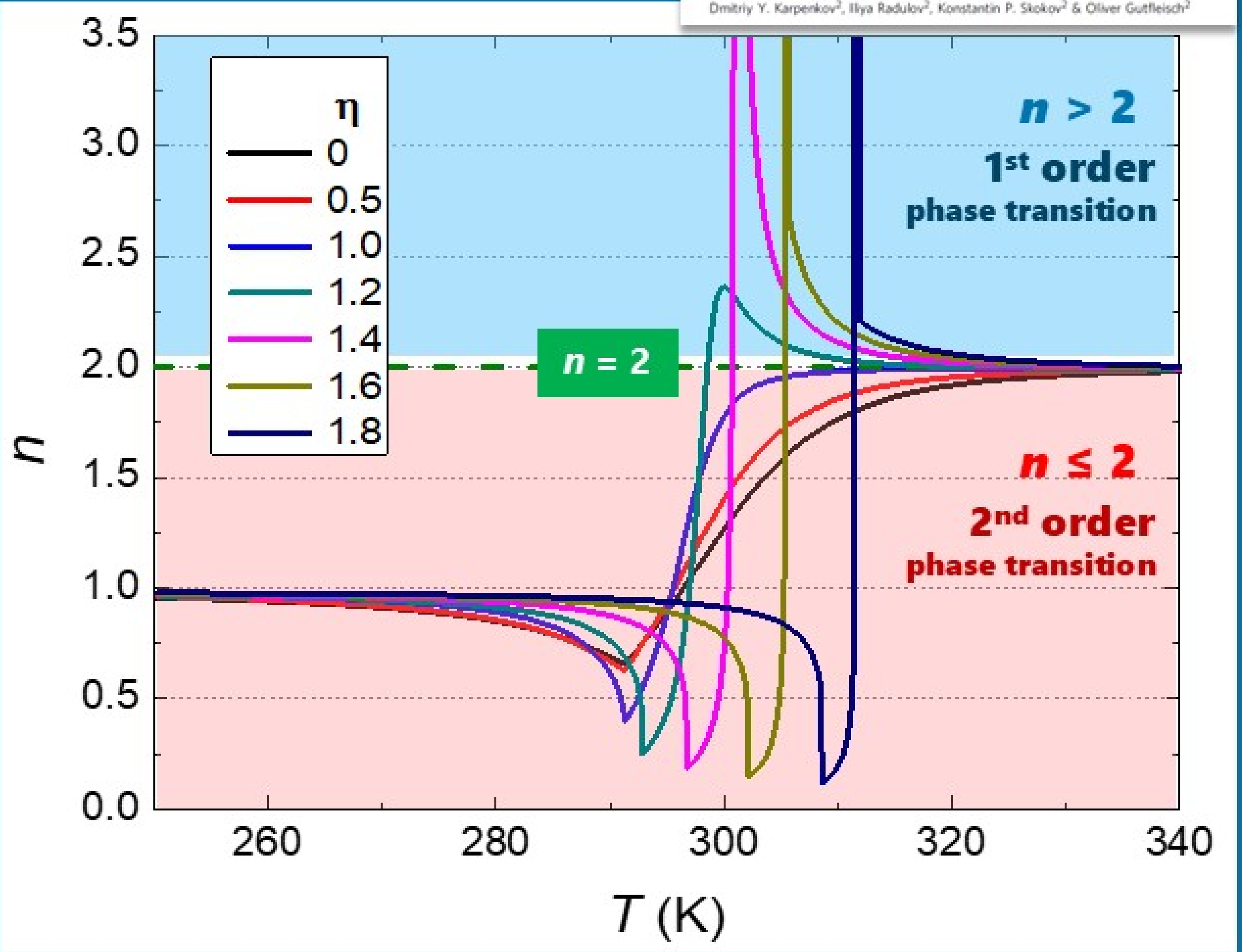
$$n = \frac{d\ln|\Delta S_{isothermal}|}{d\ln H}$$

Bean and Rodbell model (order of phase transitions)

for $0 \leq \eta < 1$: **2^{nd} order phase transition**

for $\eta = 1$: critical point

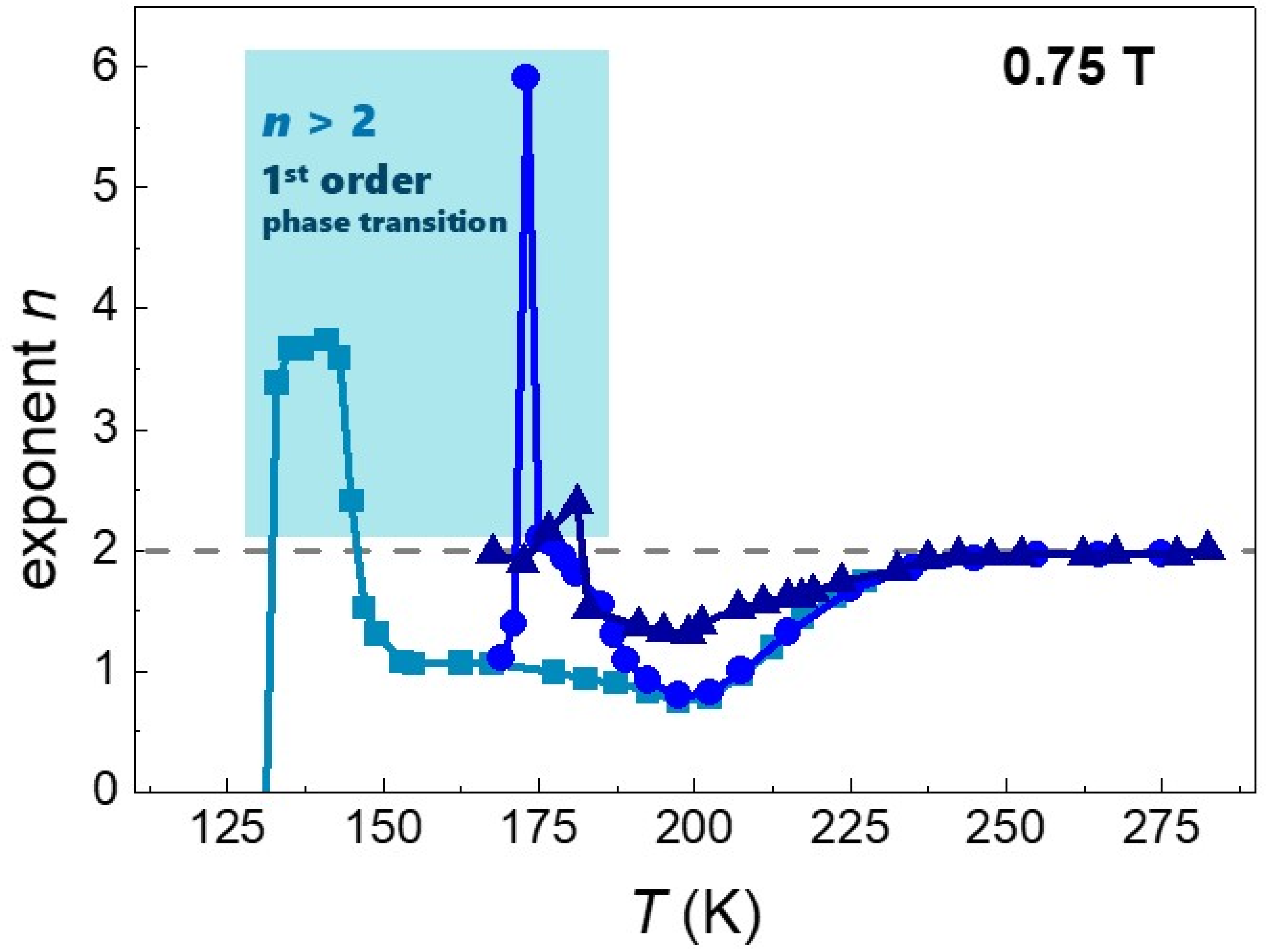
for $\eta > 1$: **1^{st} order phase transition**



JY Law et al.,
Nature Communications 9 (2018) 2680

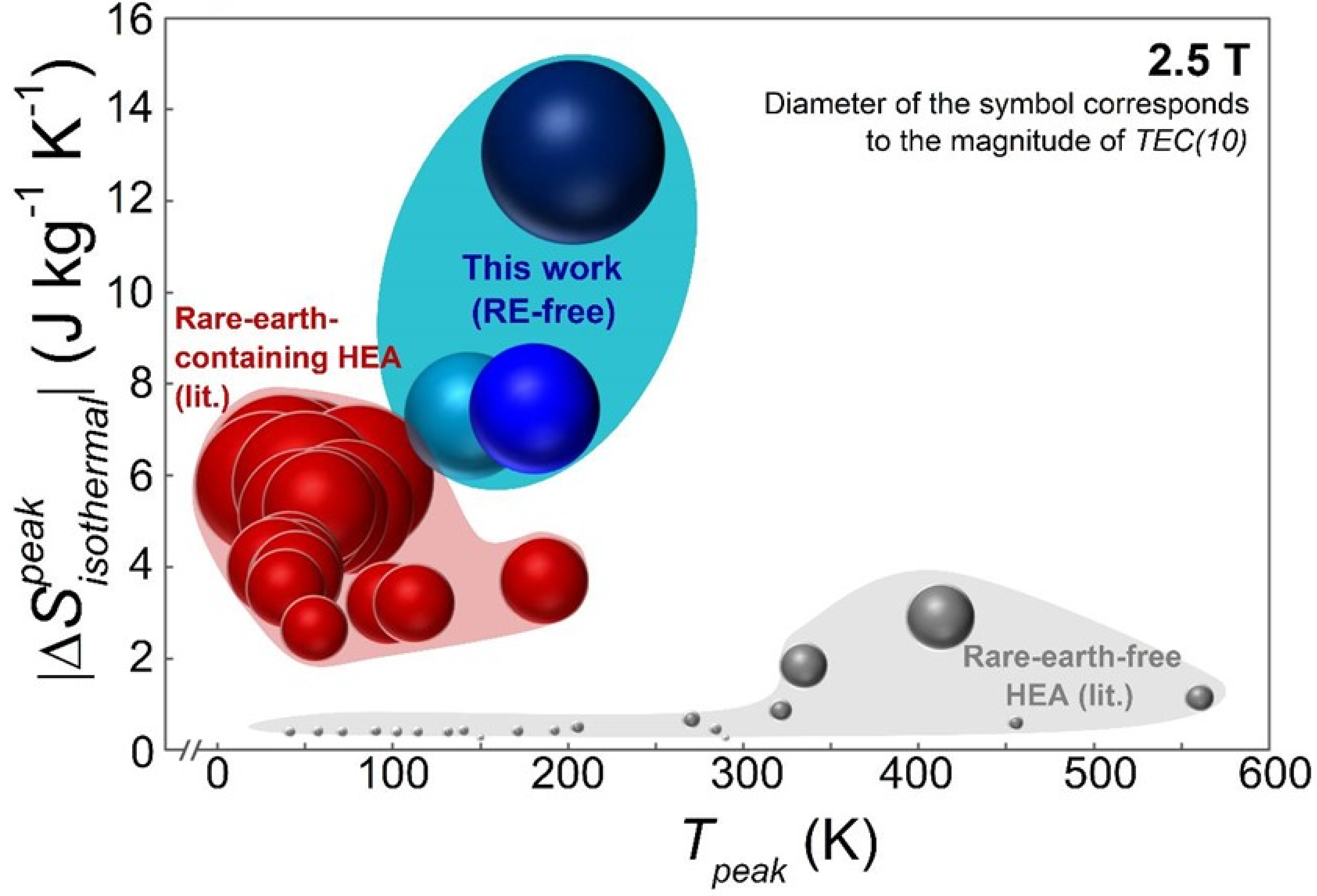
Fe-Mn-Ni-Ge-Si

order of phase transition

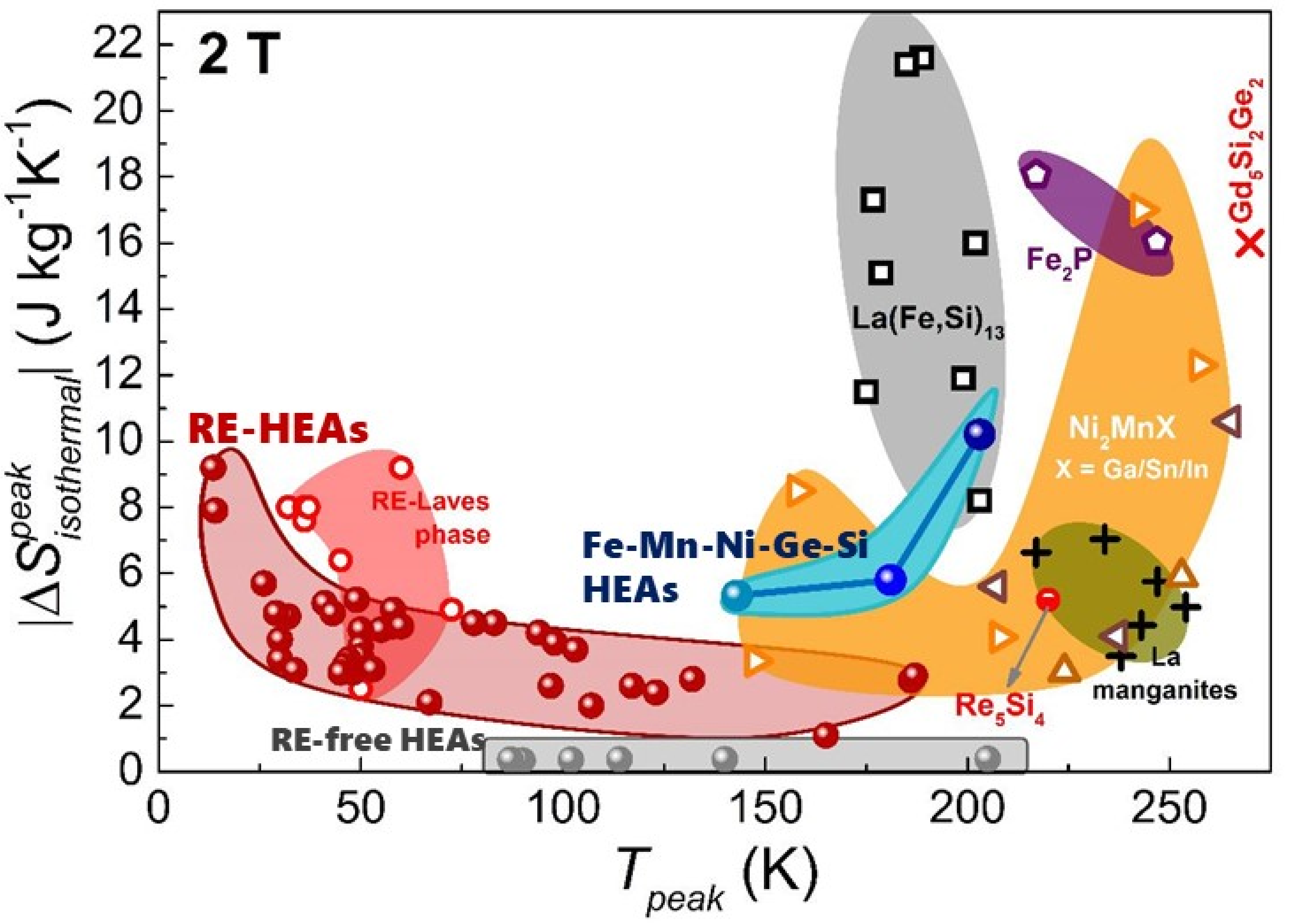


Fe-Mn-Ni-Ge-Si

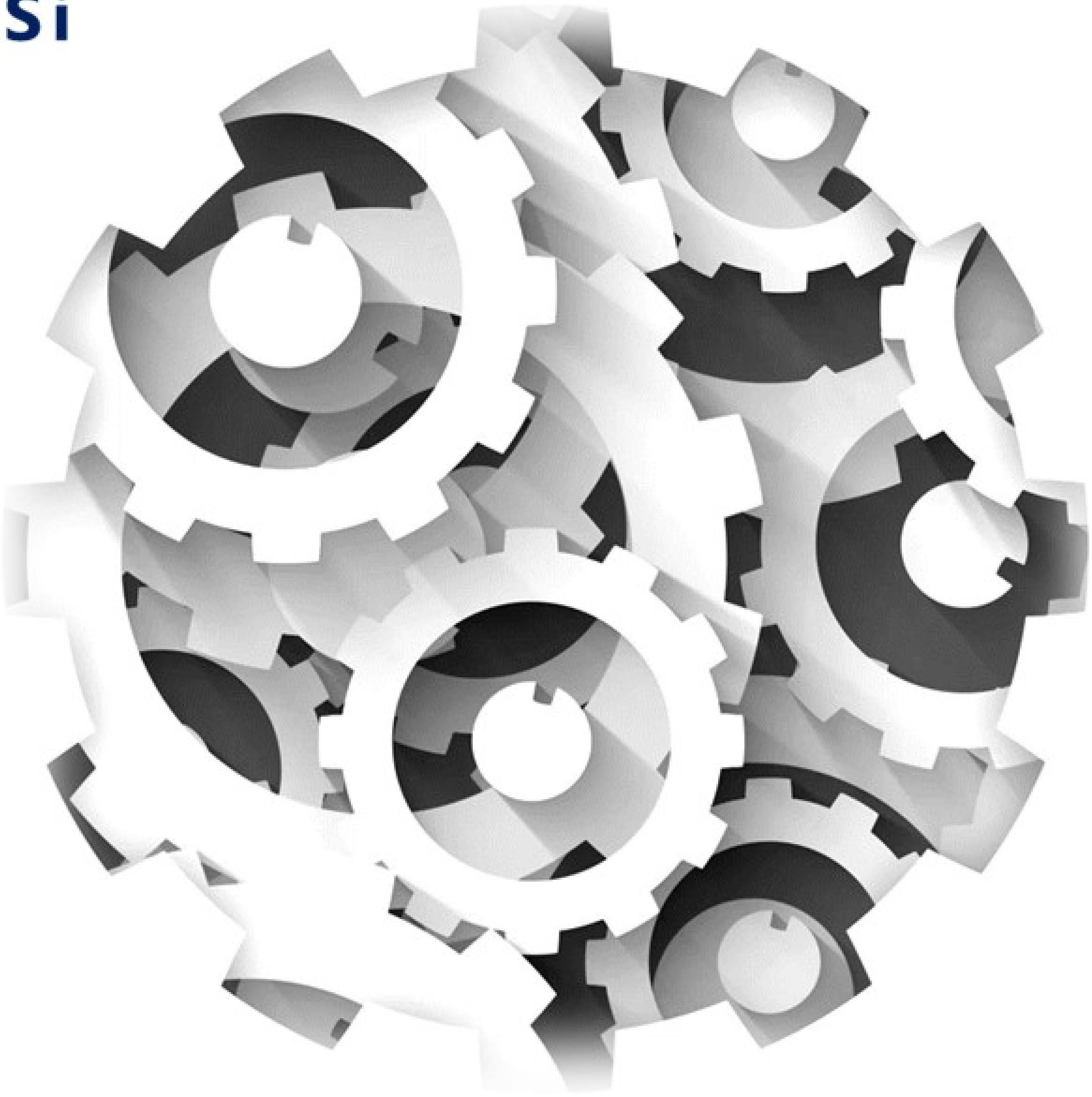
How it compares with literature HEAs



and what about with conventional MCE materials



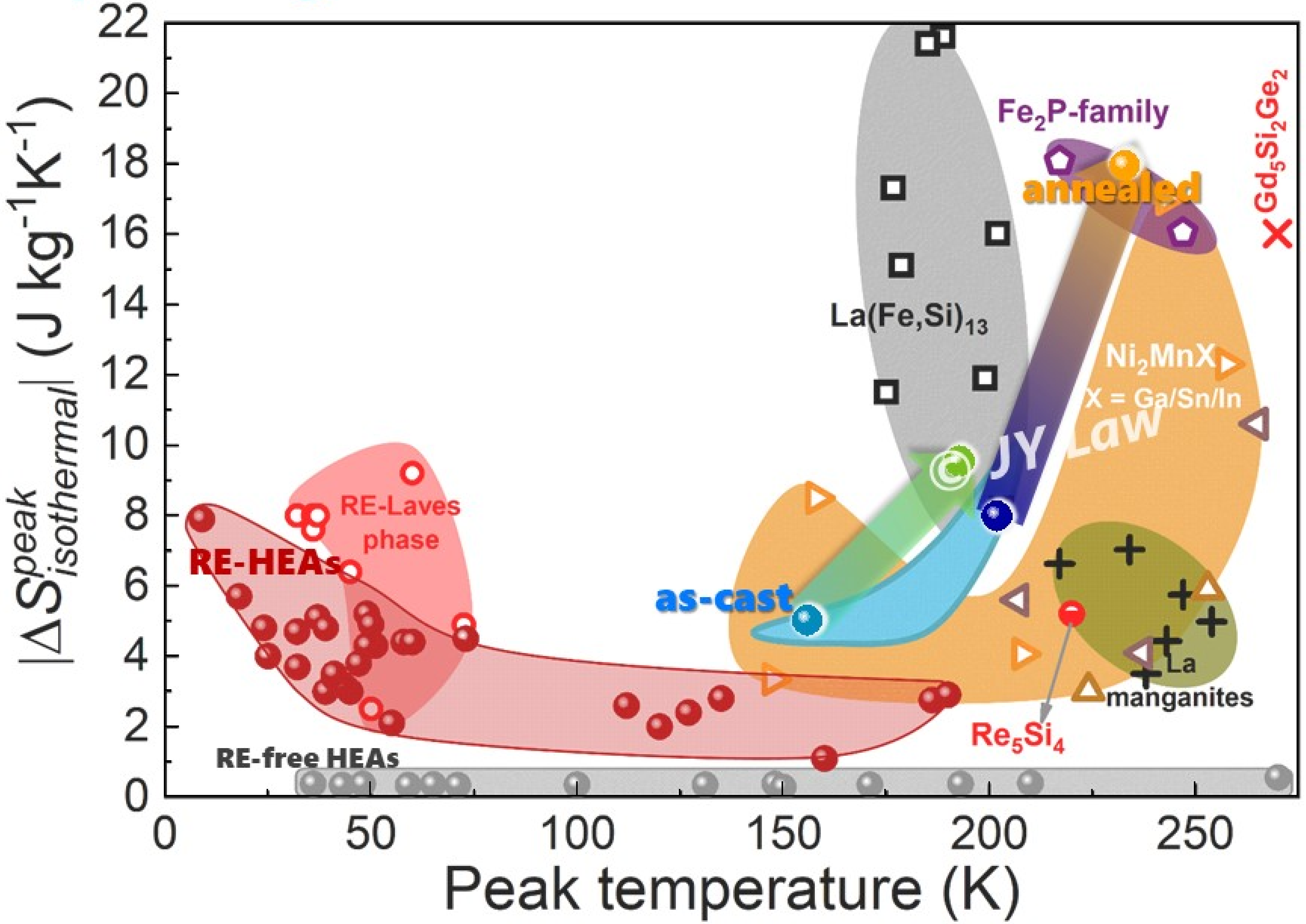
Fe-Mn-Ni-Ge-Si



brought us
to further
works

Fe-Mn-Ni-Ge-Si HEAs

and compare again




Find the HEA
Sweet Spot
when designing
compositions and tuning for
desired properties
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