

Magnetocaloric high-entropy alloys: prospects and challenges

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Thank you



Club Español de Magnetismo

Magnetotactic Bacteria:

Biological Nanorobots and Their Suitability for Cancer Therapy

by M. Luisa Fdez-Gubieda and Ana García-Prieto

University of the Basque Country, Spain

(graphics by José Manuel Barandiarán)



I am a professor at the University of Oviedo in Spain. I lead a multidisciplinary team made up of physicists, chemists, engineers, and biotechnologists who are dedicated to exploring and developing and applying nanomaterials and magnetic sensors in the life sciences. In addition, I am the Editor-in-Chief of the Magnetic Society section of the IEEE Access journal and an Associate Editor of IEEE Magnetics Letters.

In November 2022, the IEEE Magnetics Society Administrative Committee established the WIM Subcommittee under the Membership Committee to improve the development and integration of WIM in the Society.

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.

Click to join the mailing list to receive news about events and job opportunities and to participate in discussions about female researchers in magnetism.

Ever since I first attended an Intermag WIM networking event, I have been hooked; it broadened my perspective in many ways, for example, by giving me the confidence to request professional advice from a colleague or to be nominated for an invited talk. I was a young researcher at that time and had the chance to learn that senior researchers were approachable people who were honest about themselves.

I gained self-confidence from it, which helped me expand my network of contacts.

That is why I am so motivated and happy to serve as the WIM subcommittee chair, and I hope to live up to the standards of the women who have preceded me in this role.

I hope to see you in Sendai at Intermag 2023 to enjoy a worthwhile conference and an exciting WIM networking event!



Thank you CEMAG family for your support in my Editor role in IEEE Magnetics Society Newsletter

Newsletter
November 2022

Newsletter of the IEEE Magnetics Society
Volume 62 | Issue 4 | November 2022
Editor: Jia Yan Law

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IEEE MAGNETICS

IEEE Magnetics Society Newsletter



which was an effective way to promote discussion.

Another factor that was very critical for us was the hybrid path. We had to make sure that our families, many of us haven't experienced that type of situation since the COVID outbreak, yet we asked people to travel to the US from abroad. Travel was different, and rules were changing constantly. Therefore, in December 2021, Jayaan and I set off for a conference in Charlotte, NC, where we had to travel internationally. In those circumstances, we were among the very few overseas attendees. That conference confirmed my previous hunch that we needed a COVID testing site at the conference hotel if we did not want international travellers to be put through significant stress. So, we made arrangements to make that happen in New Orleans.

As mentioned above, the only certainty that we had when planning the conference was that it was full of uncertainties. We expected around 400 attendees on-site, and that was the number of on-site registrations that we had a few weeks before the conference. Unfortunately, the Omicron variant variant coincided with the moment when people had to finalize their travel plans. As a result, we lost about 10% of the ~400 on-site audience just one week before the event. We allowed people to change their registration to online and adapted the on-site program. The Program Co-chairs did a terrific job of keeping the sessions full and avoiding empty gaps in the schedule. Like modern production systems, we had to adapt a just-in-time supply chain philosophy to all this uncertainty.



The Write Stuff:
WRITER
by Ron Constant, President Elect
IEEE Magnetics Society

Although it may look grammatically correct, some phrasing of what we write is different from our own culture. We need to understand the language, which would teach us to appreciate it. Don't hesitate to point out mistakes.

If you say that you could care less because literally, what you mean, then this article is for you.

Less Is Not More

Comparative logic can be tricky. It is easy for us half as long, but hard to be half as small or to be 10 times more expensive. The original value is 5, "10 times more" is 5 + (10 x 5) = 55, for which we would probably say it is 10 times more expensive.

The expressions "n times smaller" and "n times more" do not mean what we often expect. For example, if the size is n times smaller, it is not n times less. Instead of writing that something is 10 times smaller, it is better to say it is 10 times as large. But how hard is it to say "10 times larger" instead of "10 times more"?

"Because," "as," and "since" are often used interchangeably, but they have different meanings.

"The color of the strip changed to pink" has the intended meaning that the color became pink, or is it that the color changed after it became pink?

"The color of the strip changed to pink" has the intended meaning that the color changed after it became pink, or is it that the color changed after it became pink?

I would advise one to use "because," in preference to "as" or "since".

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(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.



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I hope to see you in Sendai at Intermag 2023 to enjoy a worthwhile conference and an exciting WM networking event!

Although it may look grammatically correct, some phrasing of what we write is different from our native English. It's always worth double-checking what we've written to make sure it sounds natural.

If you say that you could care less because literally, what you mean, then this article is for you.

Less Is Not More

Comparative logic can be tricky. It is easy for us half-asleep, but hard to be half-asleep on the other side of the equation. For example, if the original value is 5, "10 times more" is $5 + (5 \times 10) = 50$, which would probably

The expressions "n times smaller" and "n logically suspect and do not mean what we often mean by multiplication. For example, if the "smaller" would be $5 - (5 \times 5) = -45$. What we mean by "n times smaller" or "n times less" is usually $10 \times 5 = 50$, for which we would probably

Instead of writing that something is 10 times larger, its inverse is 10 times as large (but not "10 times wavelength is 10 times lower" (wrong), frequency is 10 times as high" (right)).

"Because," "as," and "since" are often used together, but they can create ambiguity. Here are examples:

"The color of the strip changed to pink" has the intended meaning that the color became pink, or is it that the color changed after being pink?

"The color of the strip changed to pink" has the intended meaning that the color changed after being pink, or is it that the color changed after being pink?

I would advise one to use "because," in preference to "as" or "since".

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Prof. Victorino Franco



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Chang



Ms. Gloria
Guerrero-
Muñoz



Mr. Zhe
Cui



Ms. Elisa
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Mr. Jorge
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Magnetocaloric high-entropy alloys: prospects and challenges

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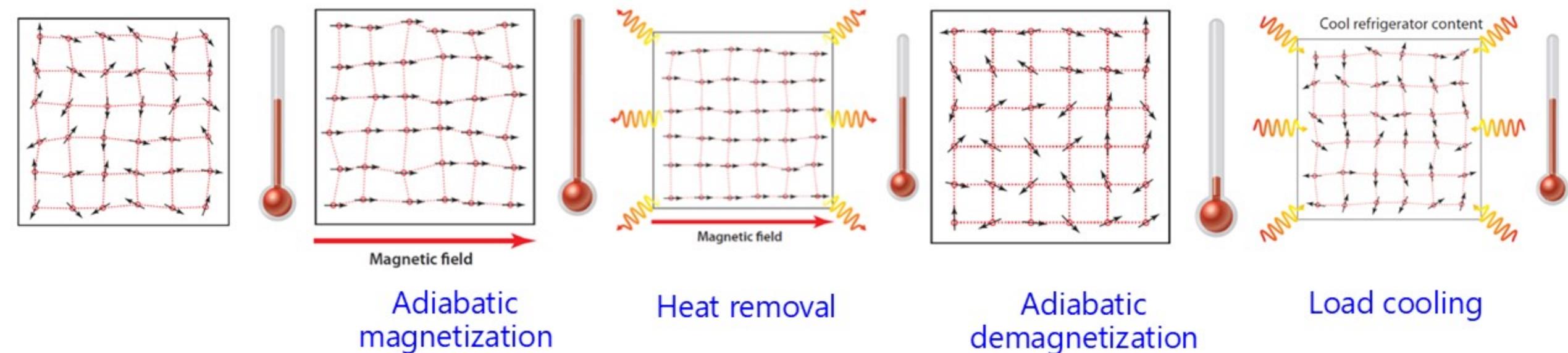
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magnetocaloric effect

refers to **adiabatic ΔT** of magnetic material
when subjected to **varying** magnetic field

4 stages of

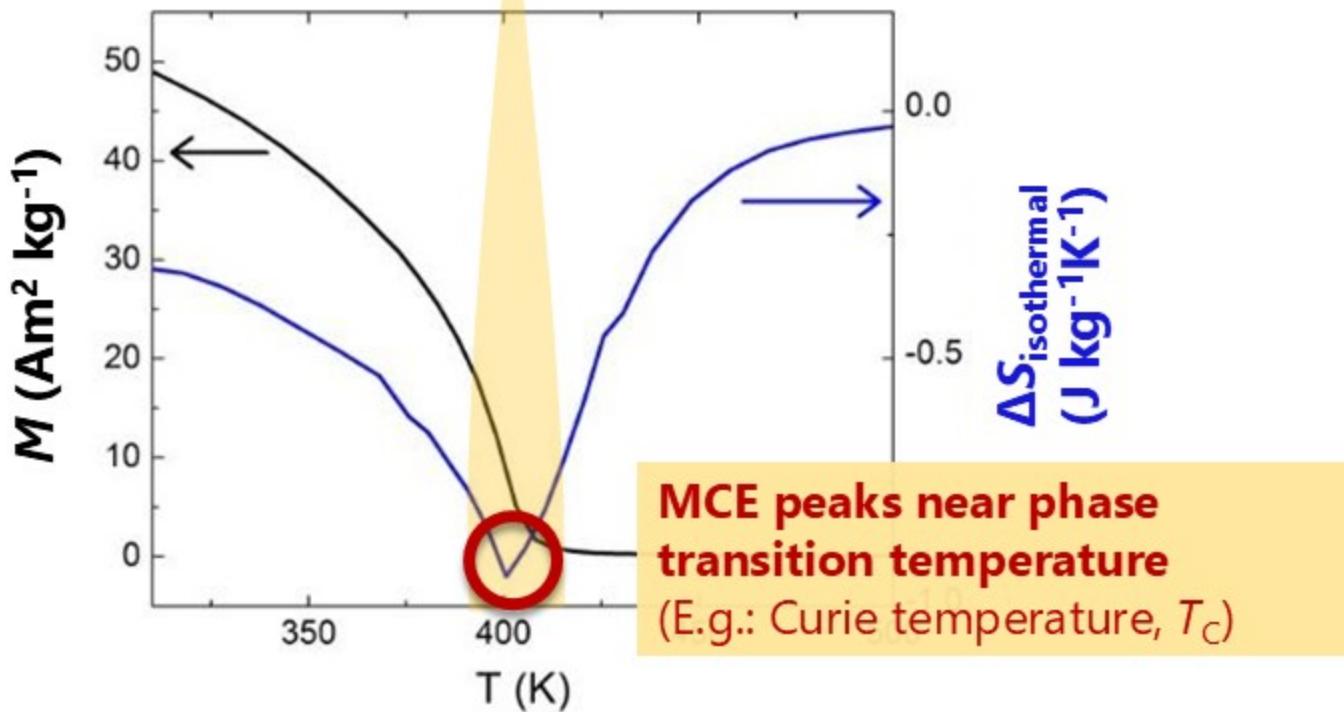
Magnetic Refrigeration Cycle



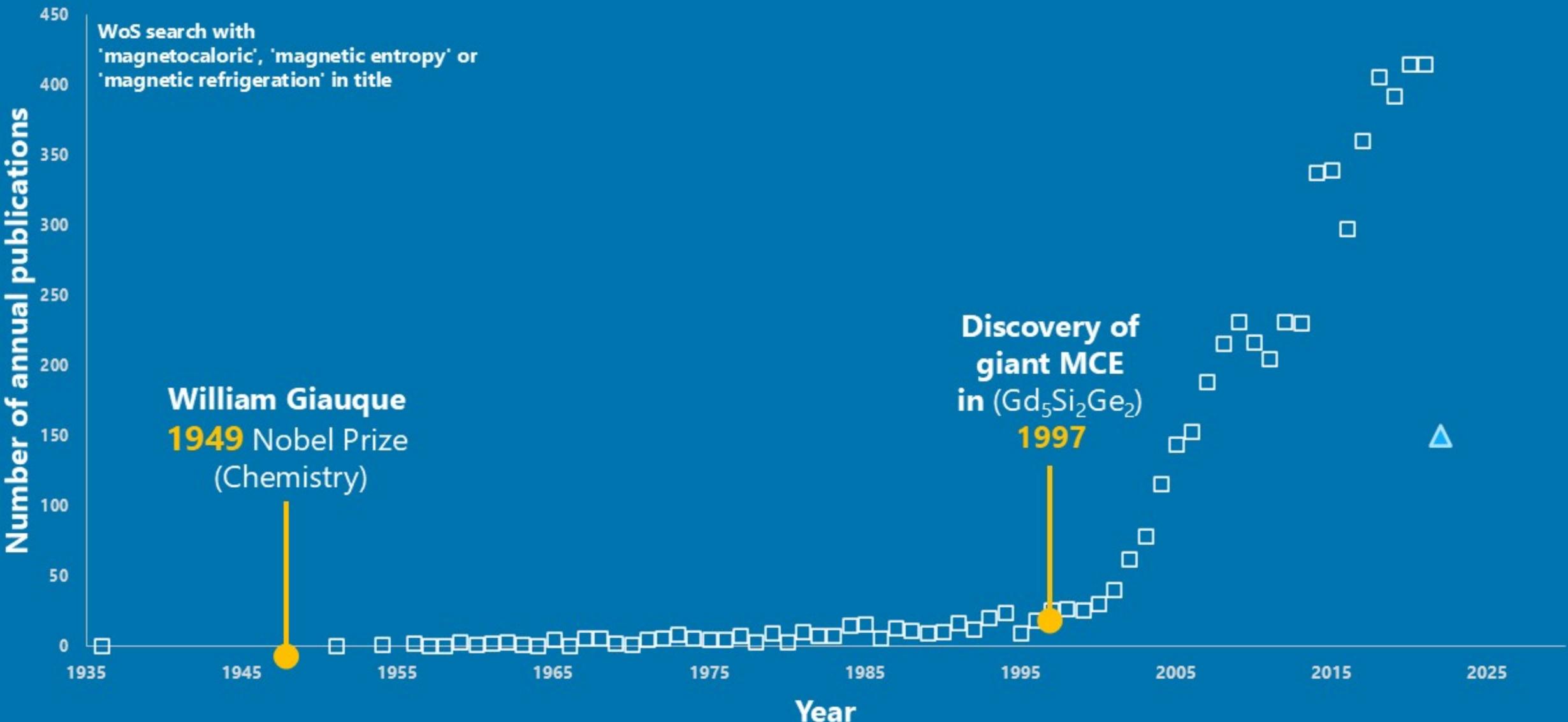
Magnetocaloric effect $\equiv \Delta T$ or $\Delta S_{\text{isothermal}}$

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

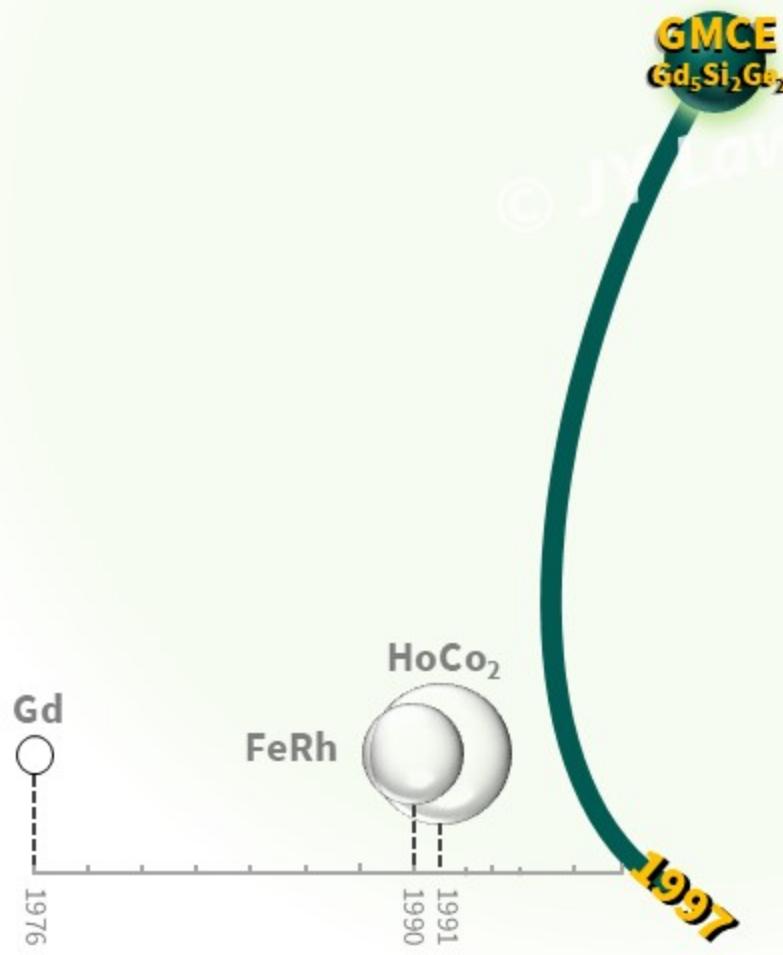
$$|\Delta S_{\text{isothermal}}| = \mu_0 \int_0^{H_f} \left(\frac{\partial M}{\partial T} \right)_H dH$$



milestones



FOMT compounds that *flourished* after GMCE $\text{Gd}_5\text{Si}_2\text{Ge}_2$

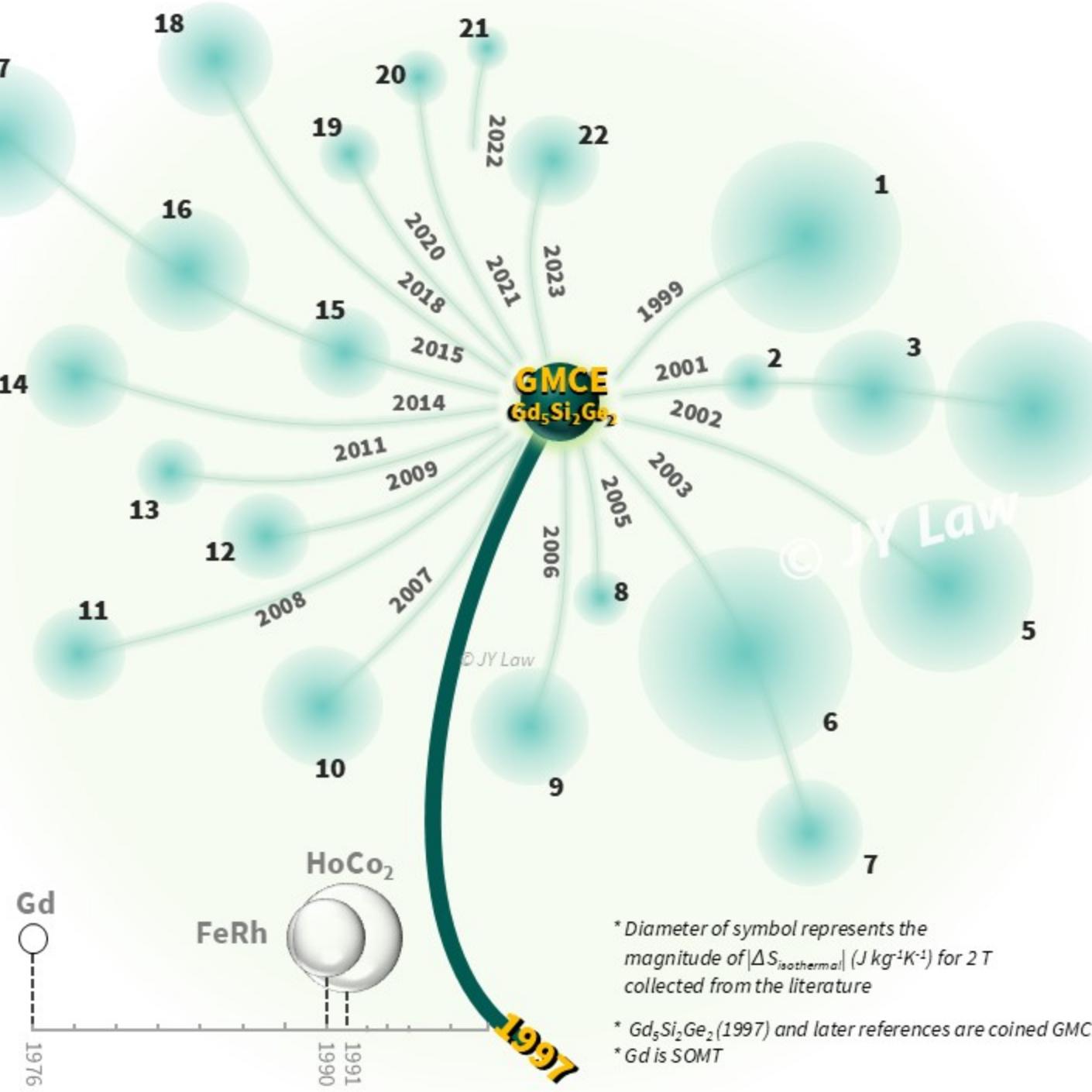
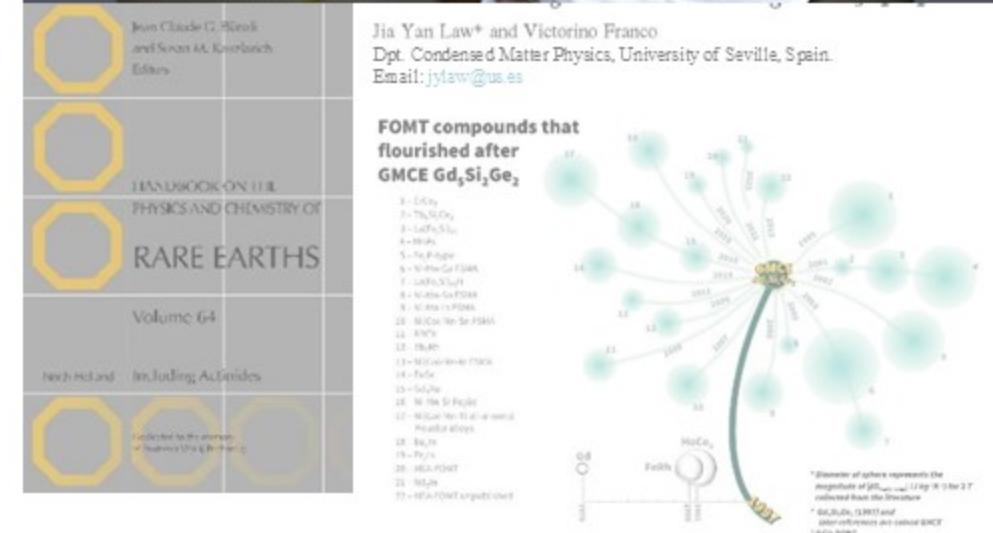


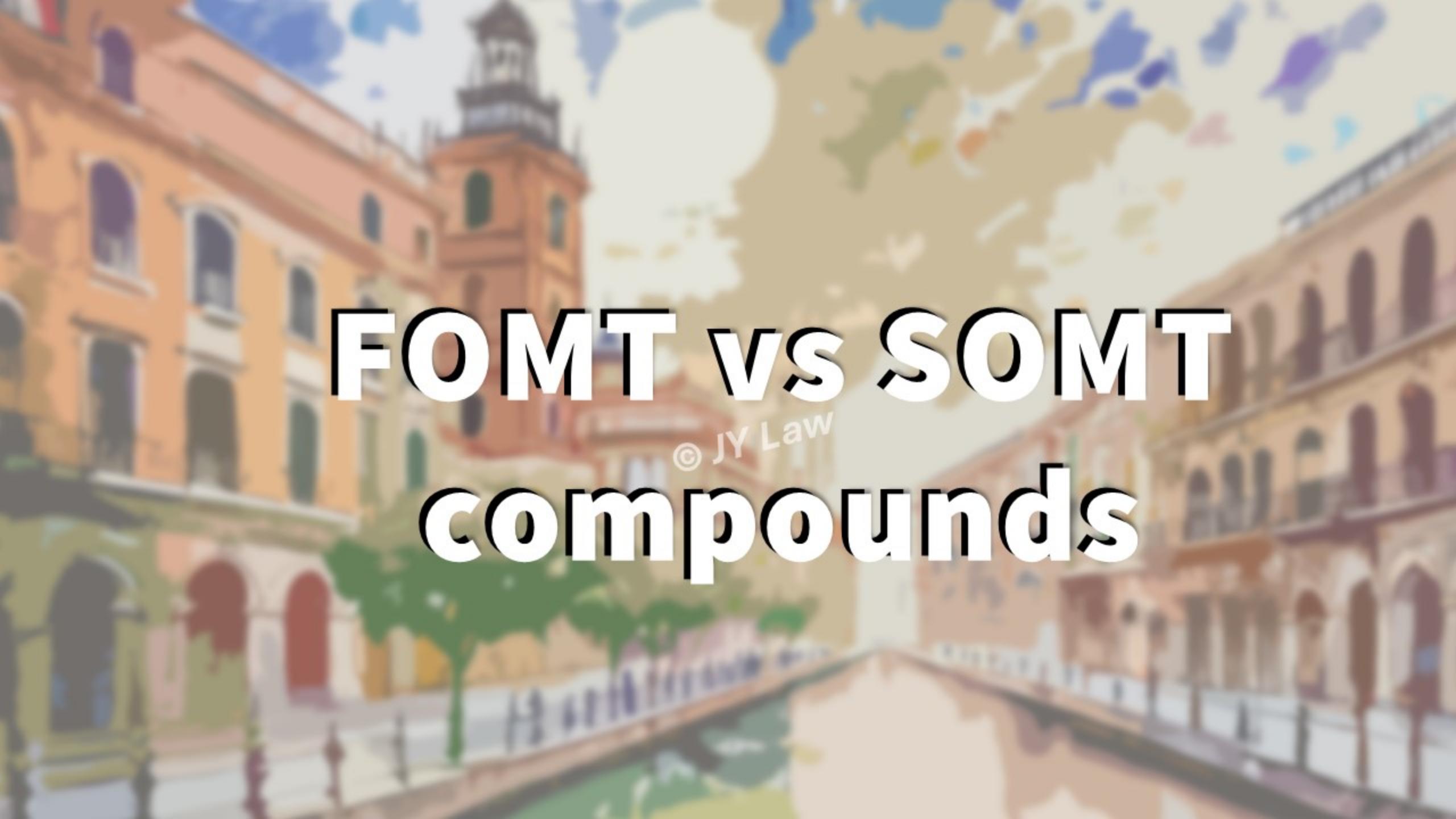
* Diameter of symbol represents the magnitude of $|\Delta S_{\text{isothermal}}|$ ($\text{J kg}^{-1}\text{K}^{-1}$) for 2 T collected from the literature

* $\text{Gd}_5\text{Si}_2\text{Ge}_2$ (1997) and later references are coined GMCE

* Gd is SOMT

FOMT compounds that flourished after GMCE $\text{Gd}_5\text{Si}_2\text{Ge}_2$





FOMT vs SOMT compounds

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MCE

- Larger ΔS_{iso}
- Narrow T_{span}
- Hysteresis
- Rate-dependent behavior

1st order

Large ΔS_{iso}
No thermal hysteresis

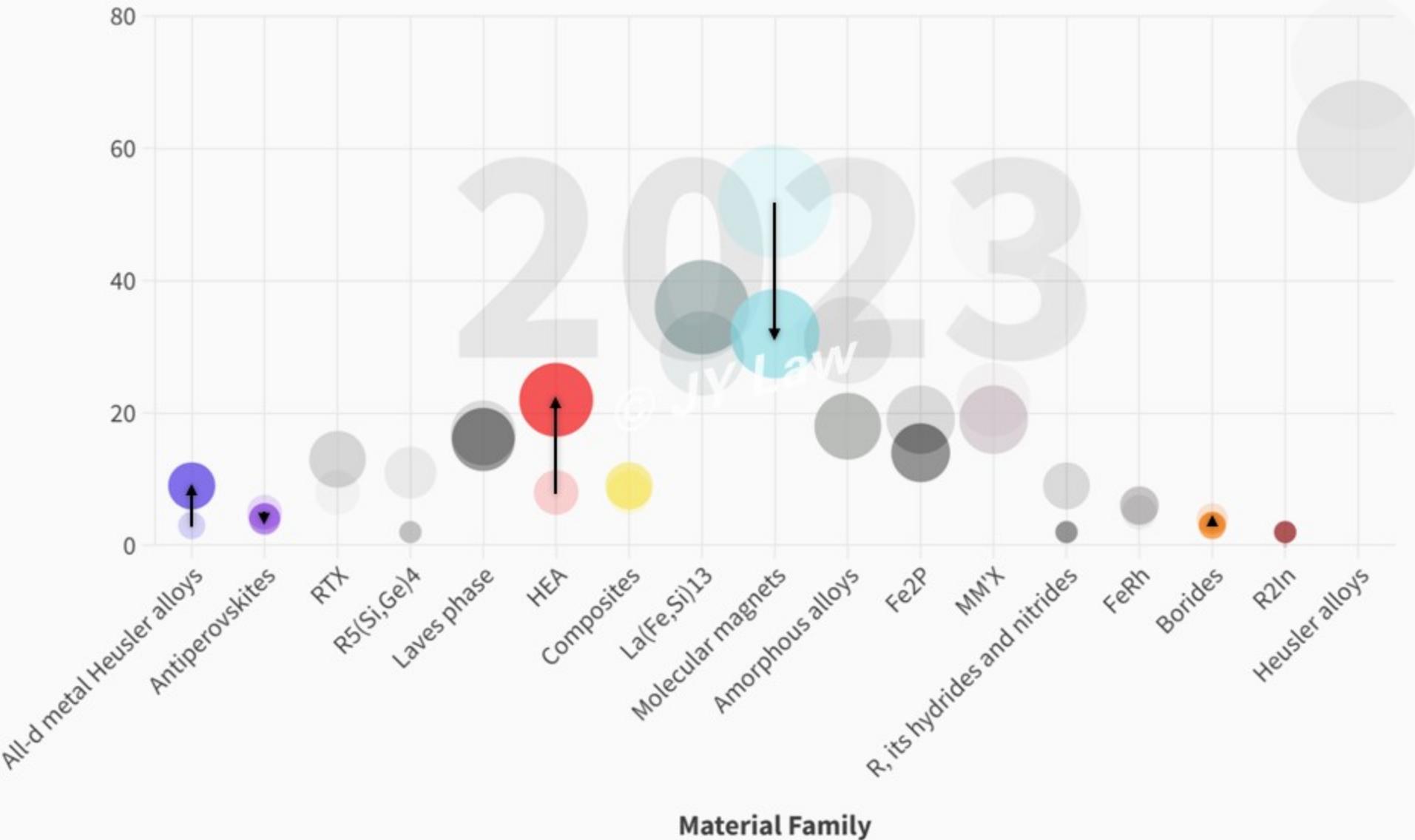
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- Moderate ΔS_{iso}
- wider T_{span}
- No thermal hysteresis

2nd order



Annual Publications (2019 – 2023)



Concept of HEA

Traditional alloys or LEA



Equiatomic HEA

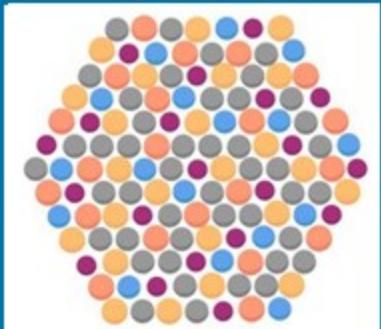
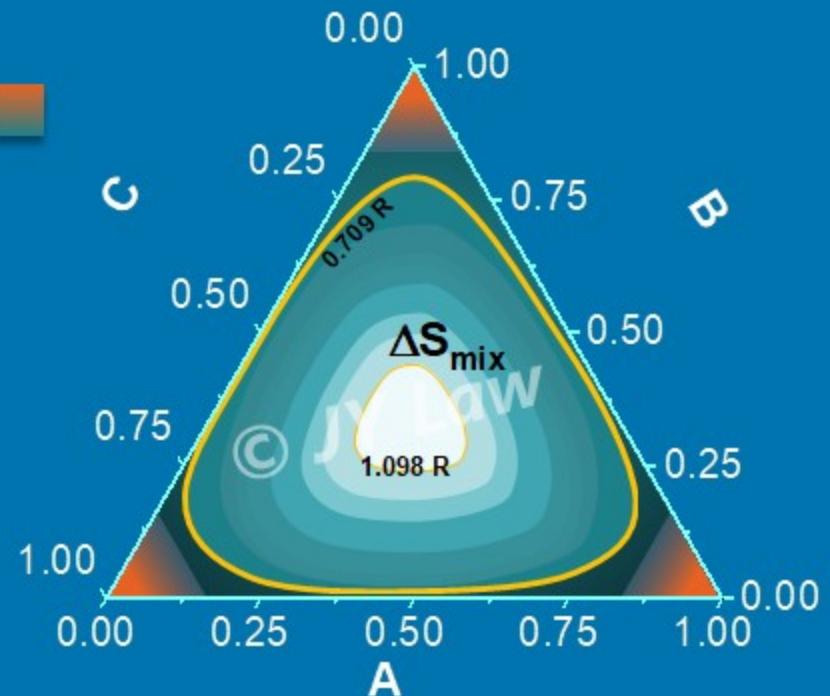


Image taken from J Mater Chem A 9 (2021) 663



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

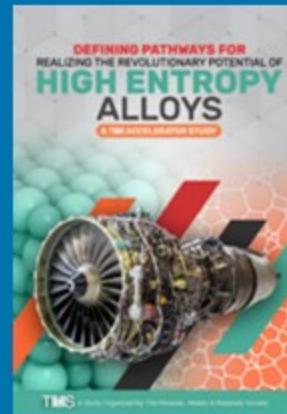
1st generation HEAs:

restricted to

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

Today

HEAs have evolved to
2nd generation...



Concept of HEA

Traditional alloys or LEA



Equiatomic HEA

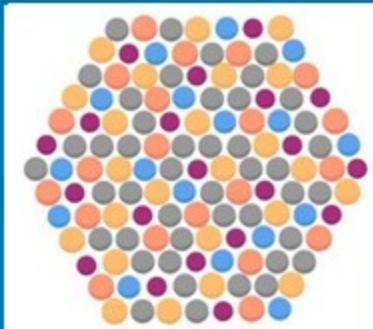
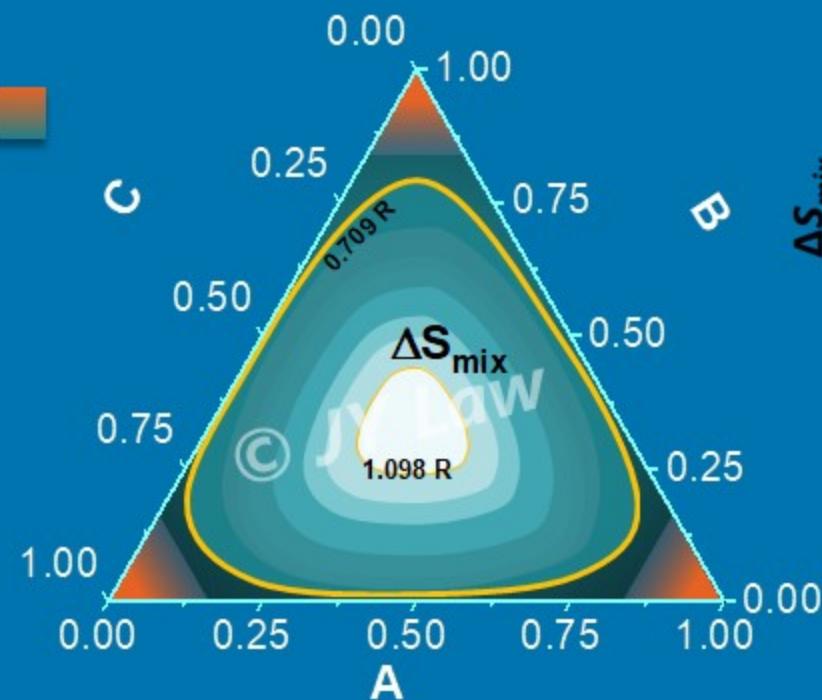


Image taken from J Mater Chem A 9 (2021) 663

multi principal elements



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

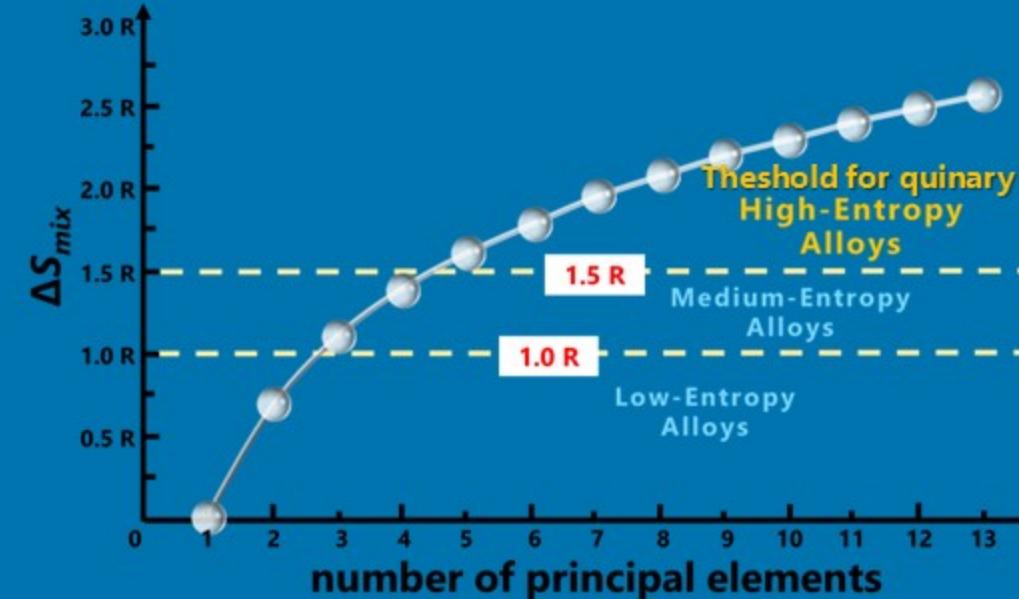
Configurational entropy of mixing

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

R = gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$)

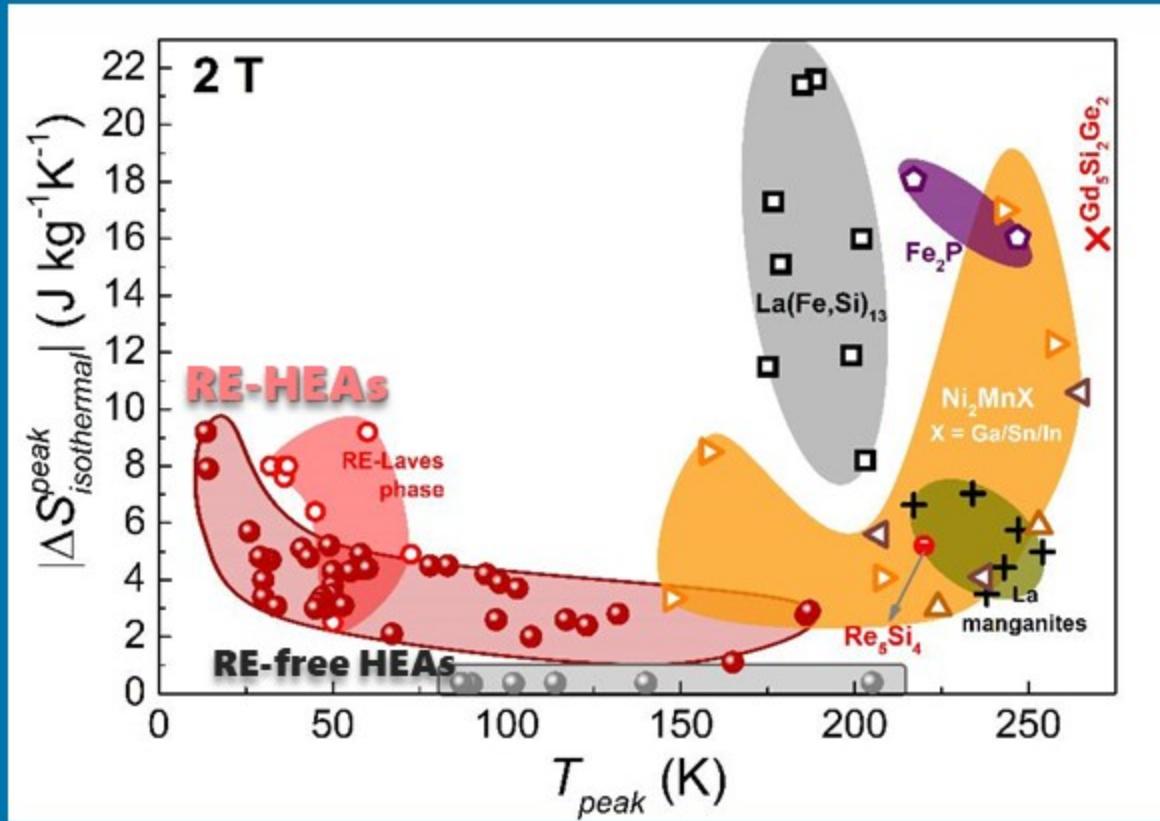
n = total # constituent elements

c_i = molar fraction of i^{th} element



HEAs are far from being competitive

compared to conventional magnetocaloric materials



HEA publications

and their different focus

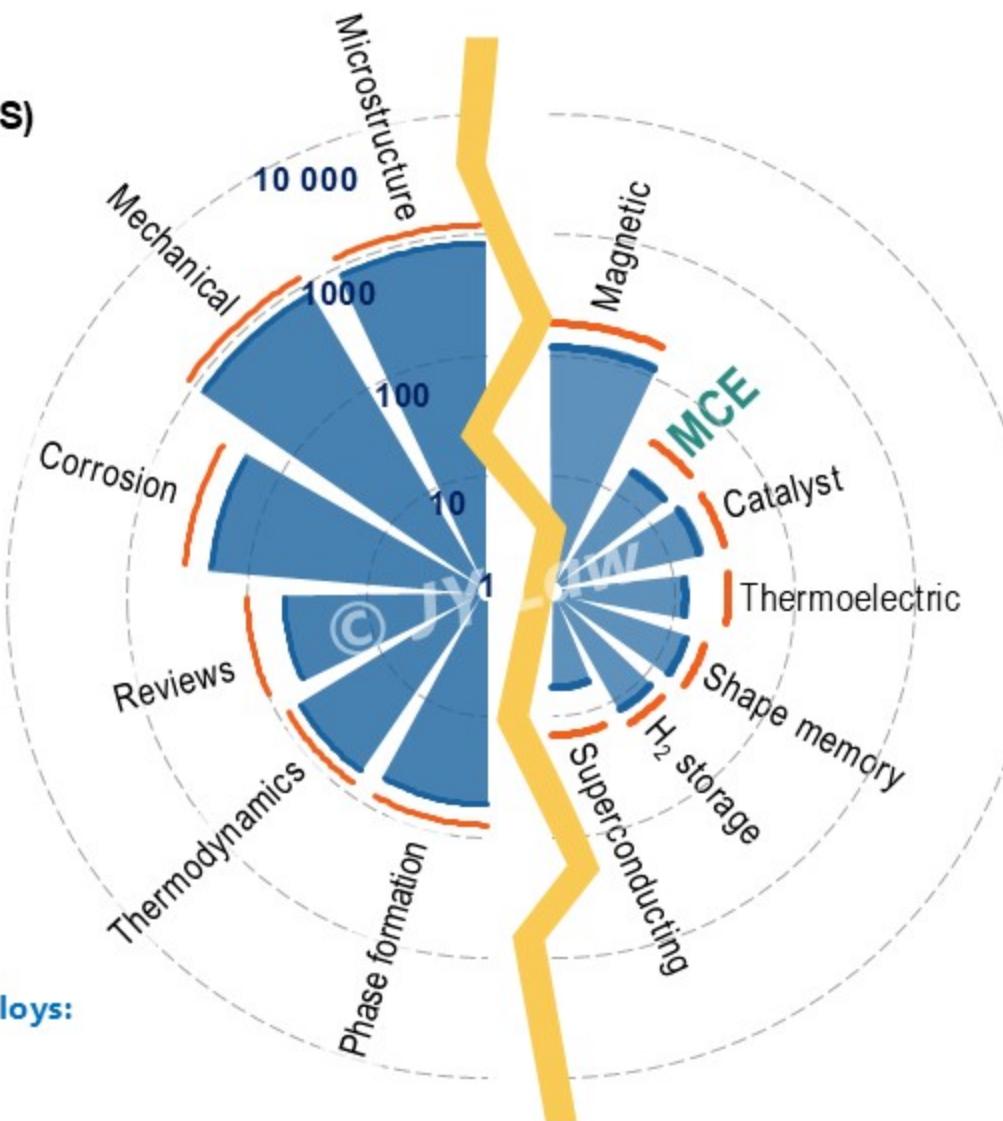


Bibliographic search (WoS)

May 2022 —
May 2021 —

structural

functional



JY Law, V. Franco,

Review on Magnetocaloric High-Entropy Alloys:

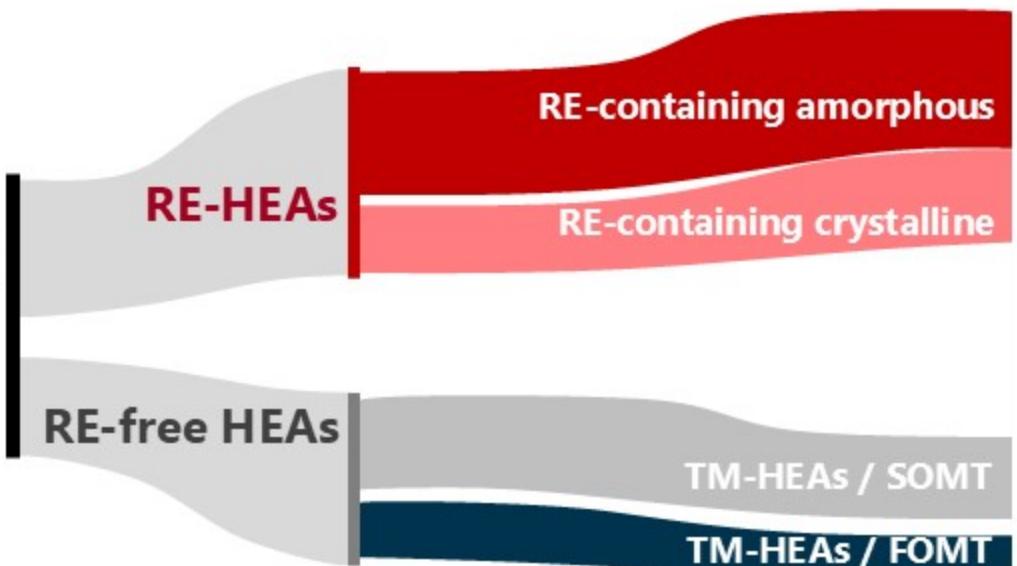
design and analysis methods,

J Mater Research **38** (2023) 37-51



Magnetocaloric HEAs

in the literature



Pushing the limits of magnetocaloric high-entropy alloys

Cite as: APL Mater. 9, 080702 (2021); doi: 10.1063/5.0058388

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Jia Yan Law[✉] and Victorino Franco[✉]

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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.

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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 $\Delta S_{\text{mix}} \geq 1.5R$ regardless of being single or multiphase at room temperature, in which ΔS_{mix} is calculated as

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

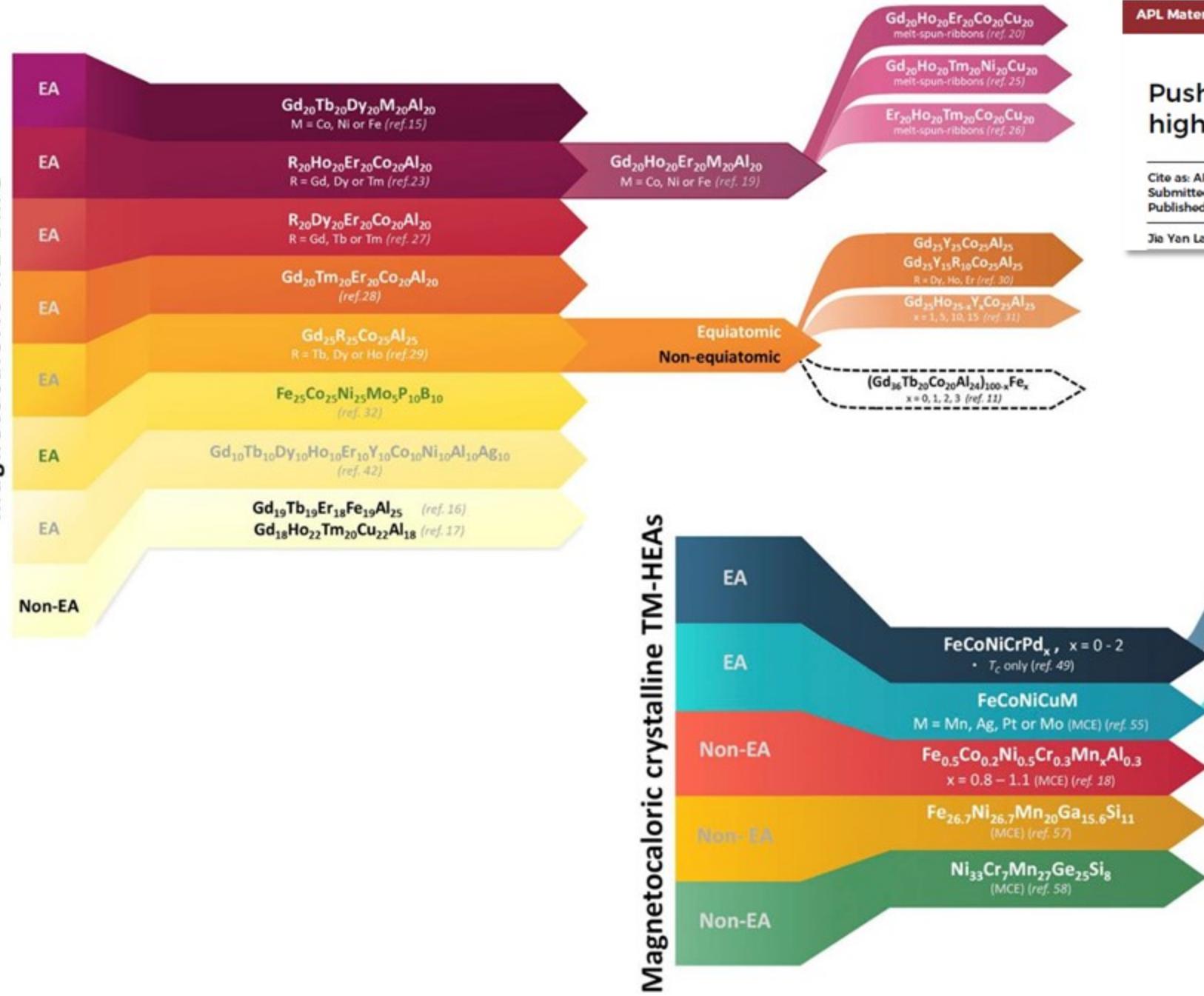
where R is the gas constant and x_i is the mole fraction of the i th element. It is worth noting that the entropy of mixing is usually simplified to exclusively consider the configurational entropy since this term dominates over the other contributions to the total entropy of mixing, which are vibrational, 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

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

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 i th major and j th minor elements, respectively.



Pushing the limits of magnetocaloric high-entropy alloys

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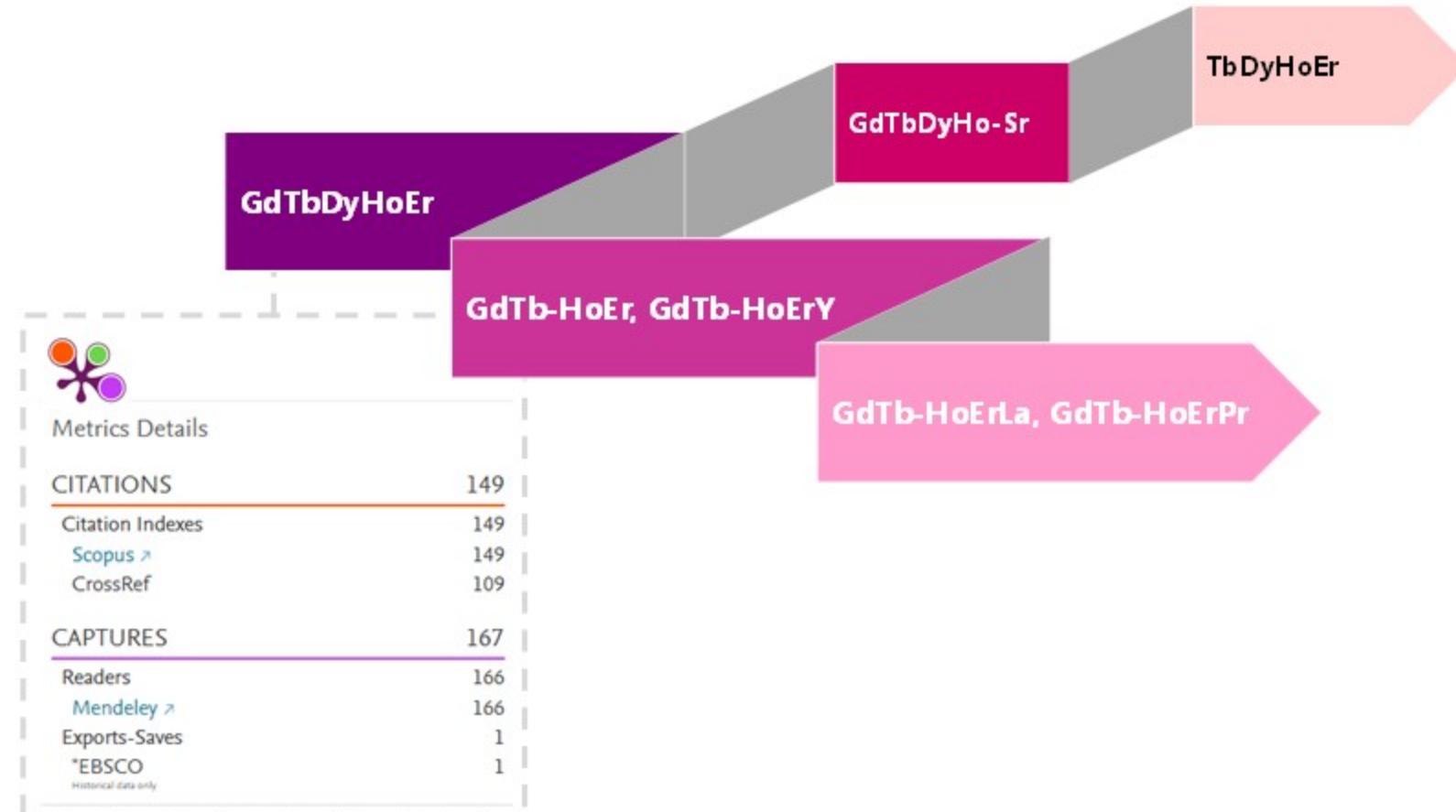
Jia Yan Law^{a1}  and Victorino Franco^{a1} 



Pushing the limits of magnetocaloric high-entropy alloys

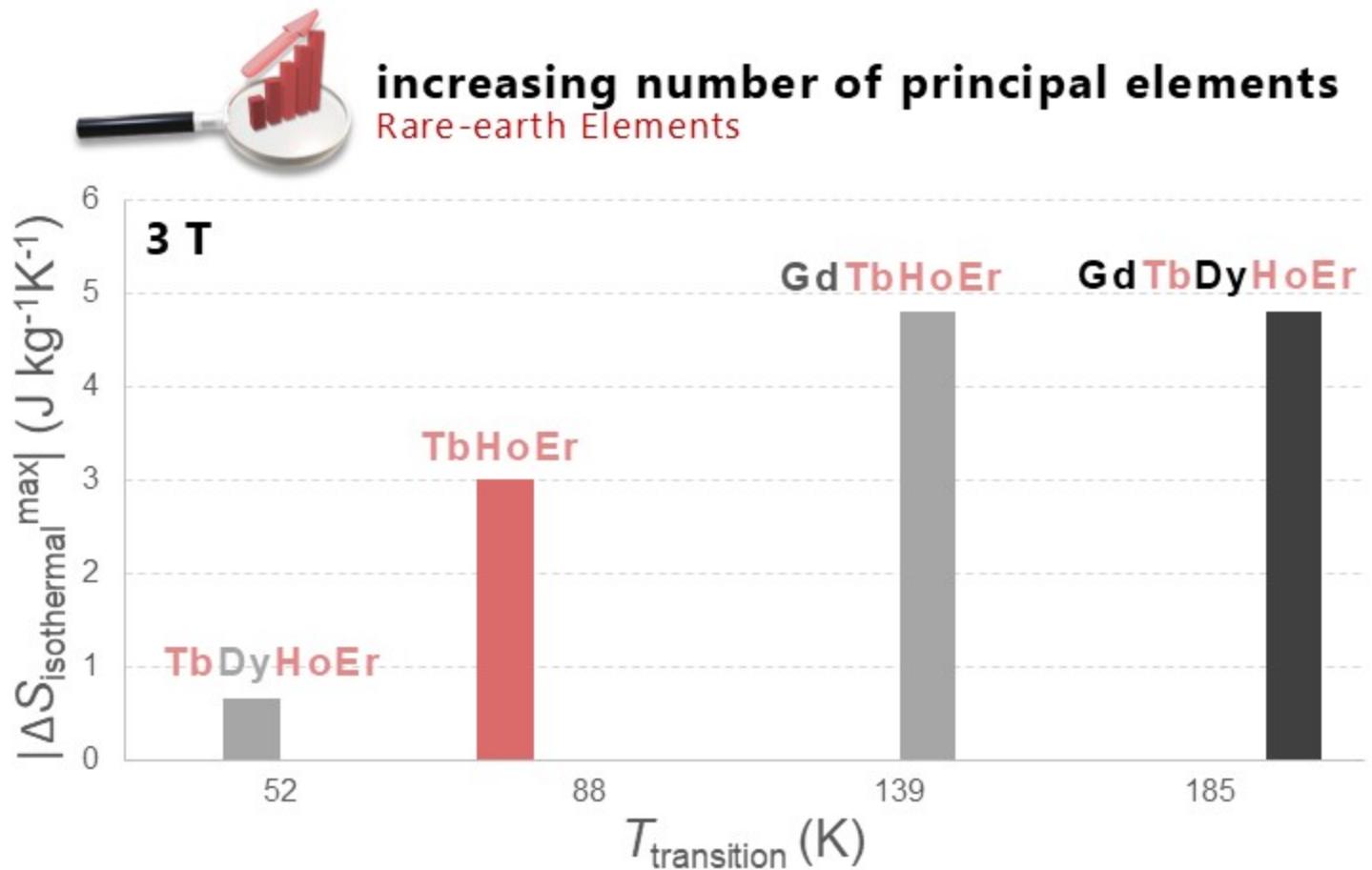
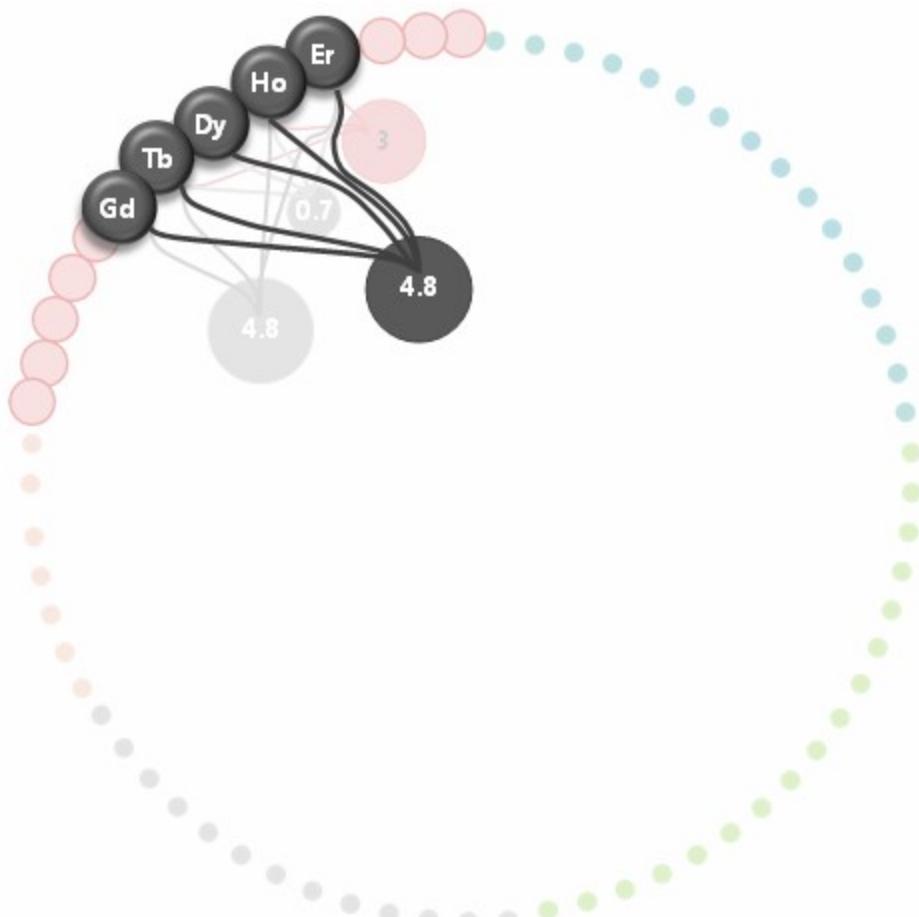
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Search strategy

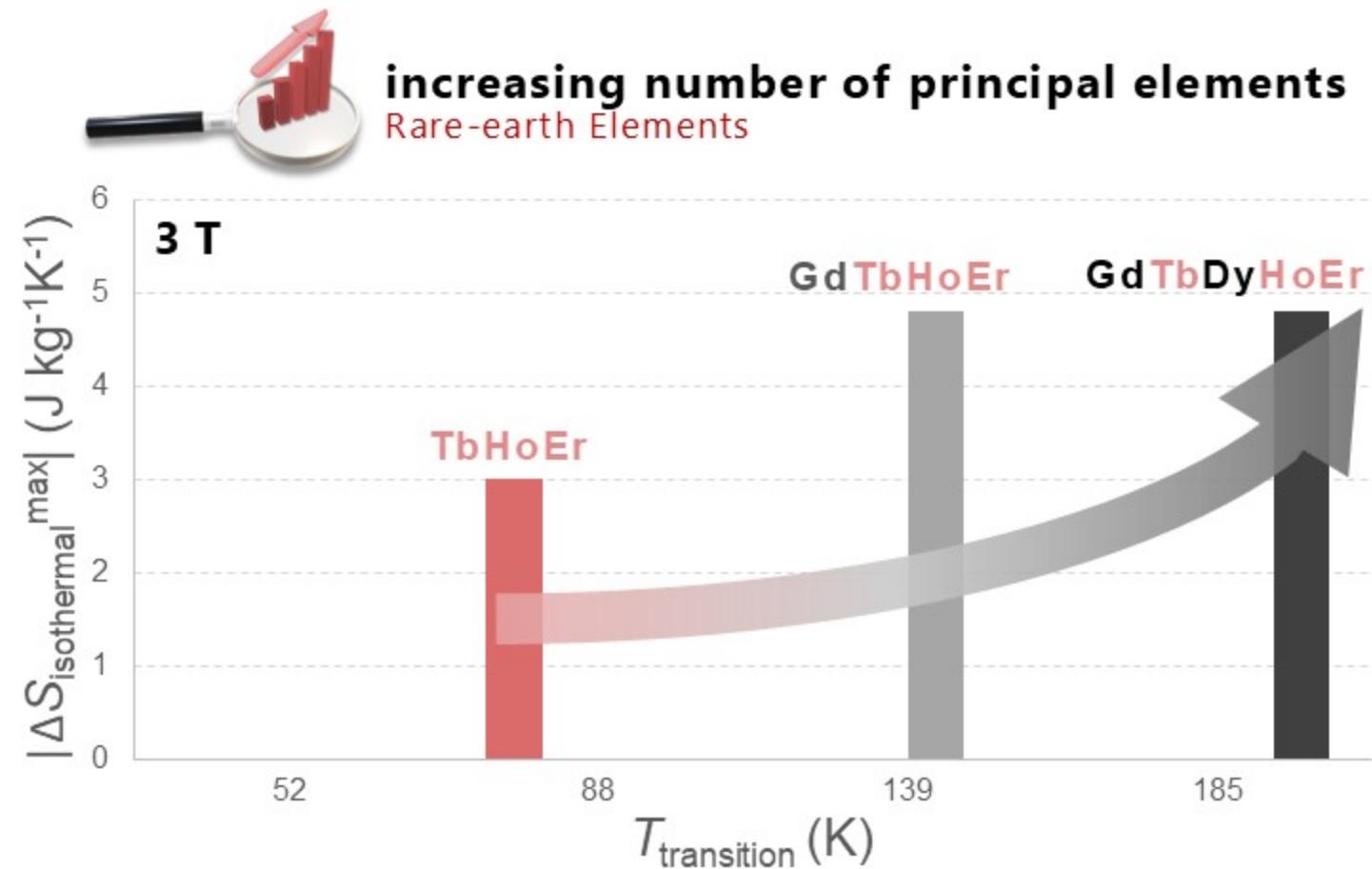
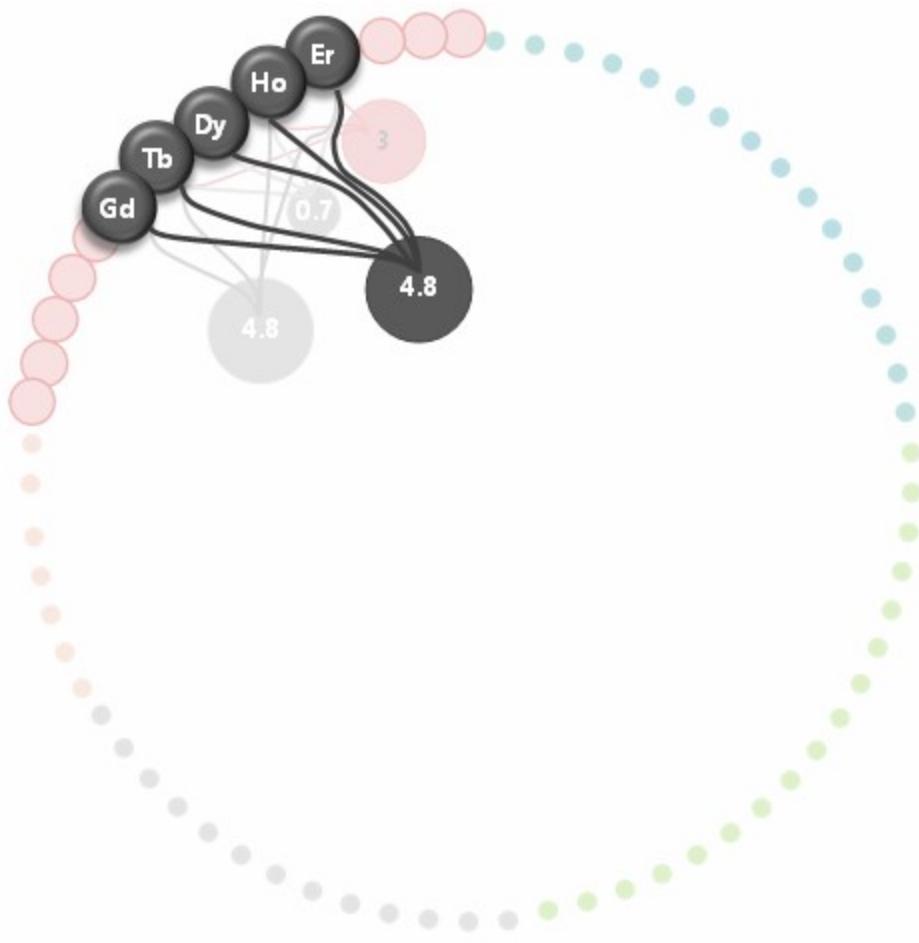
through the large HEA space



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

Search strategy

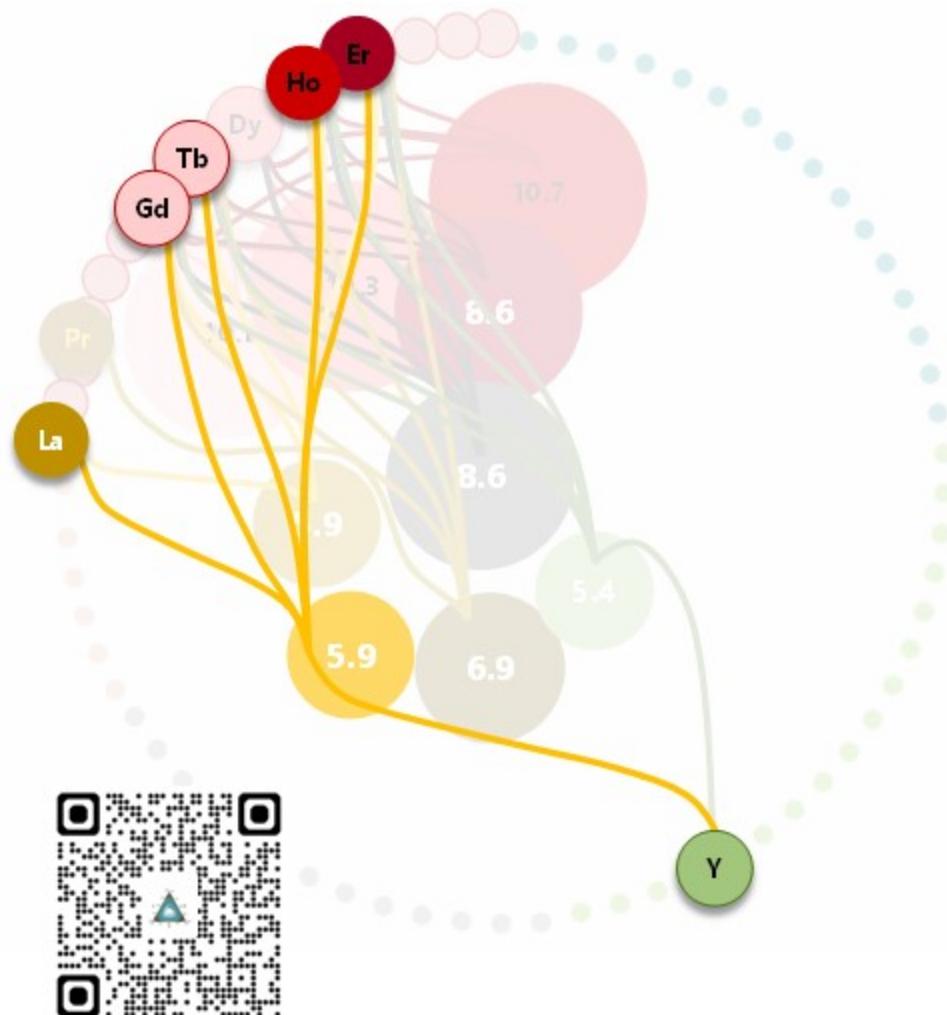
through the large HEA space



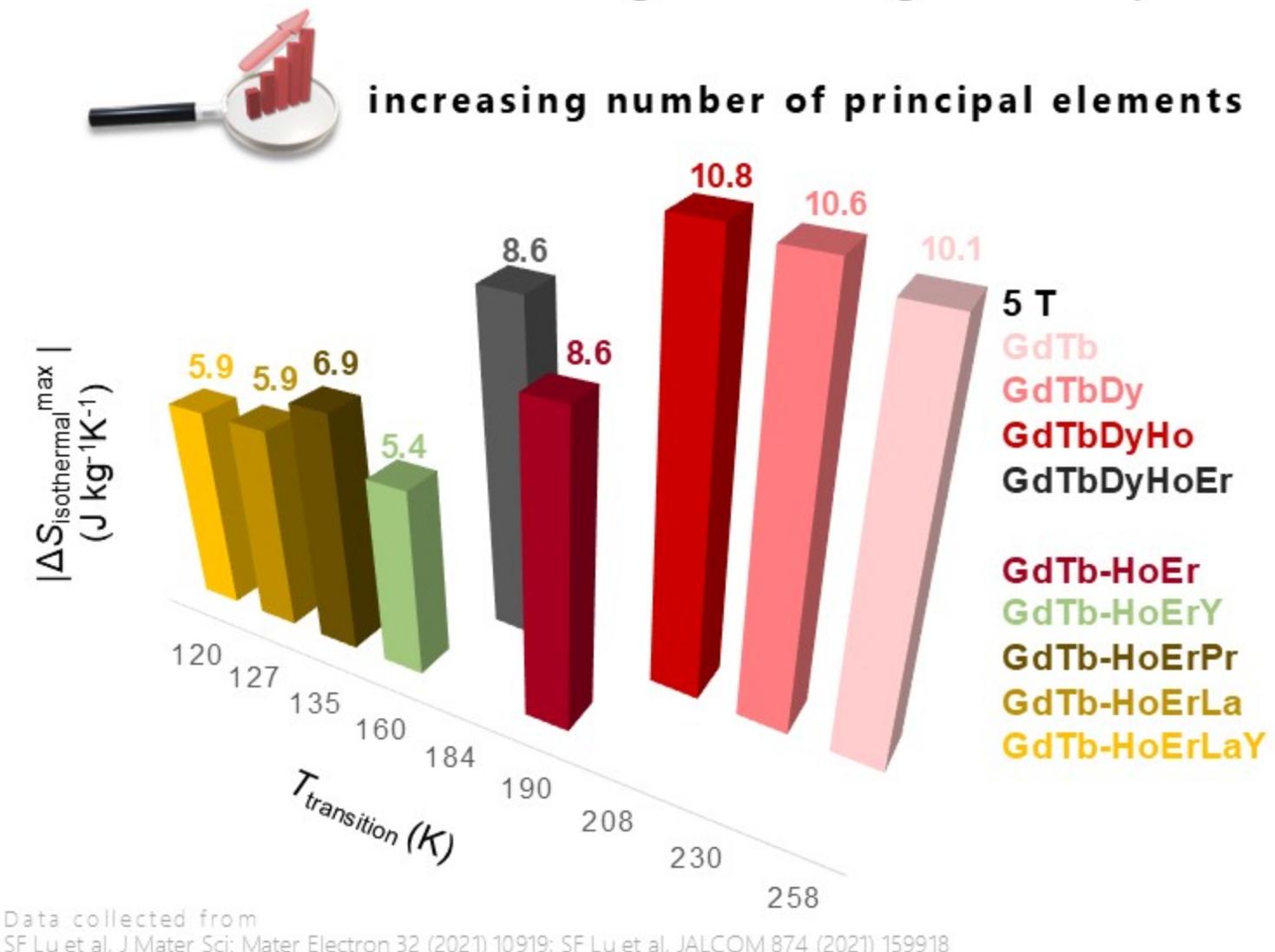
Data collected from
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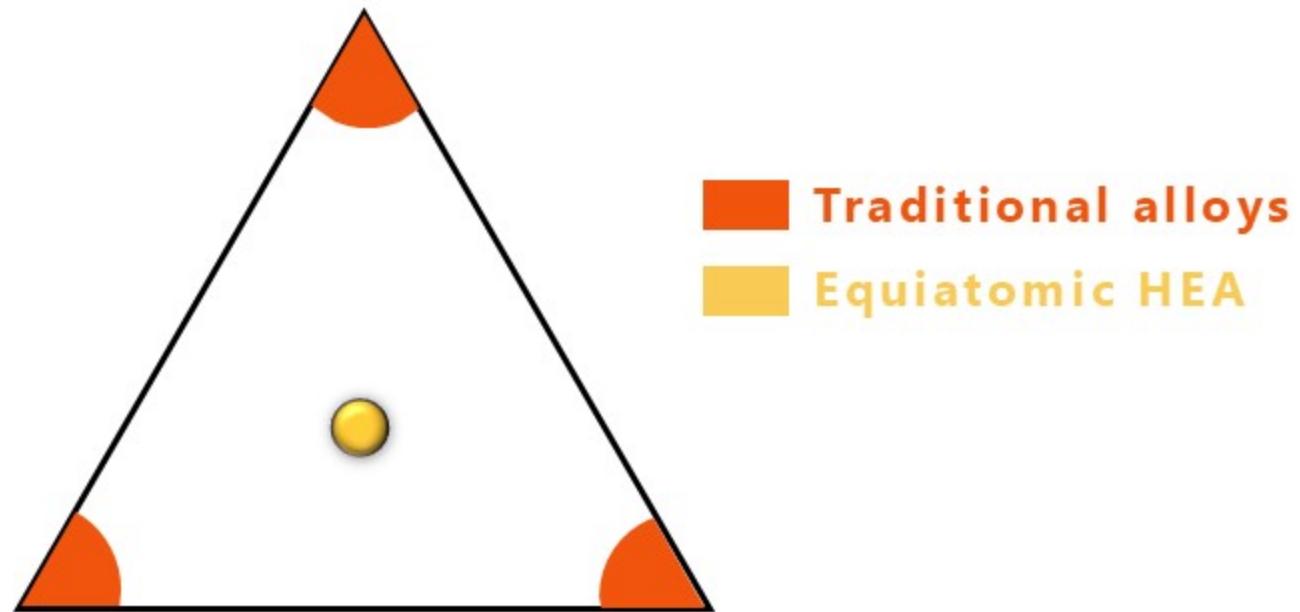


Search strategy

through the large HEA space

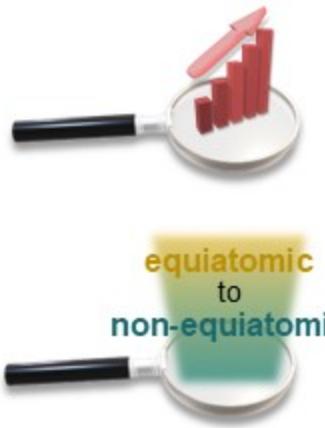


increasing number of principal elements

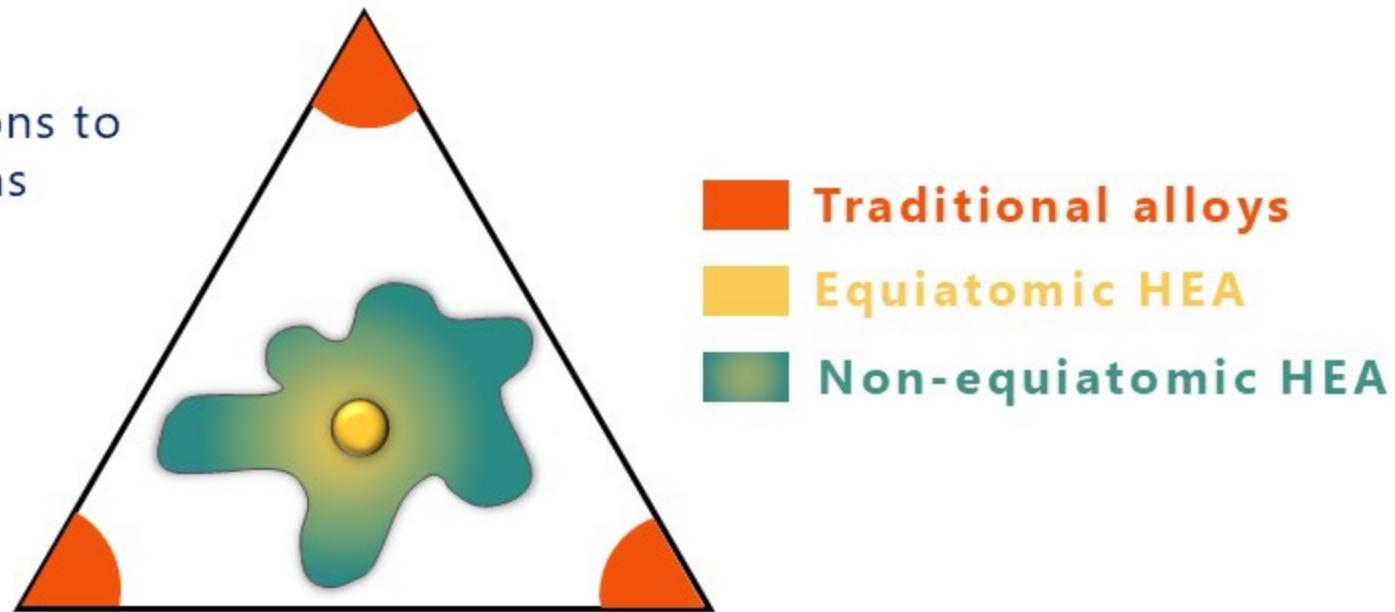


Search strategy

through the large HEA space



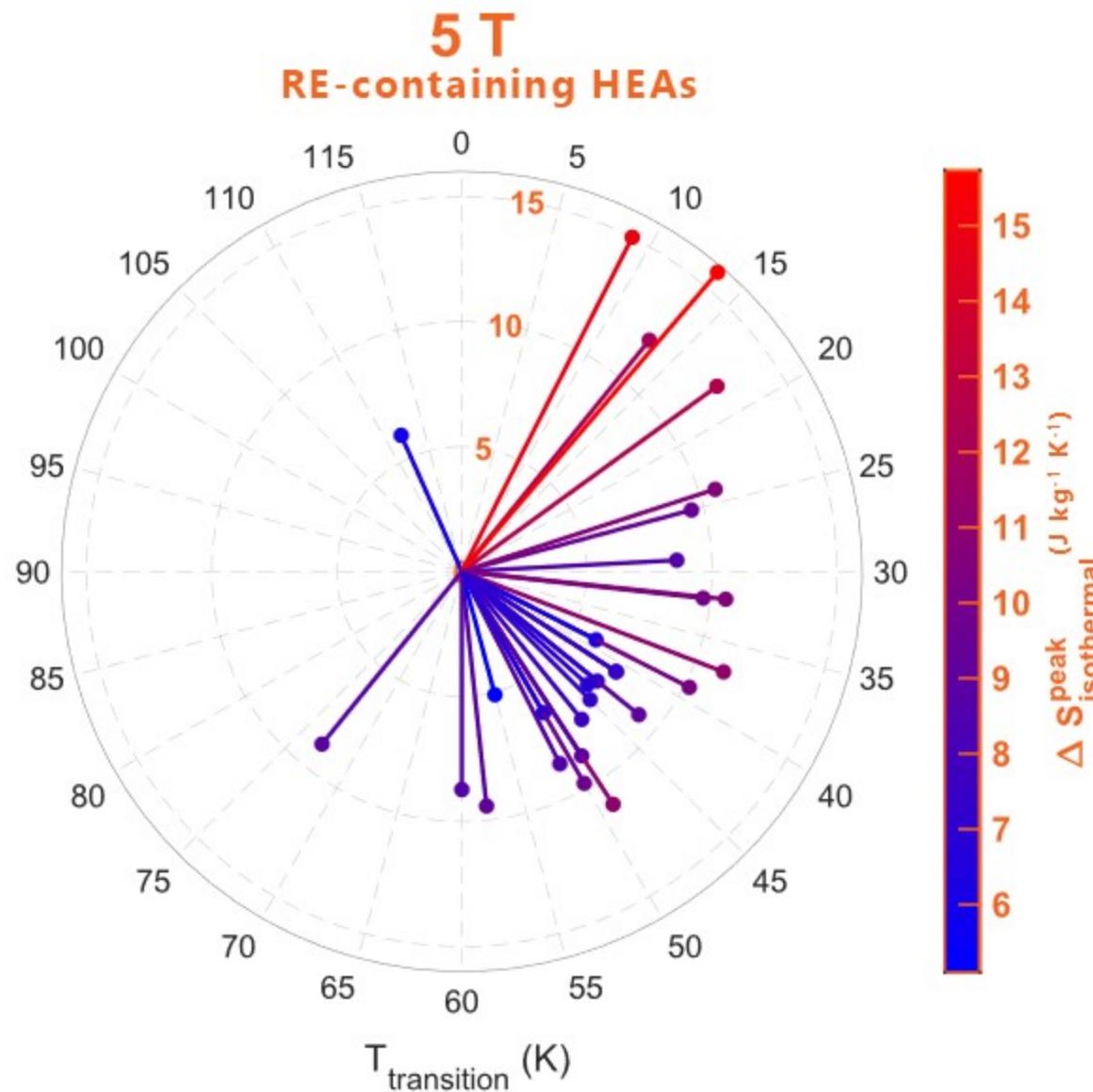
extending from
equiatomic compositions to
non-equiautomic regions



- Traditional alloys
- Equiatomic HEA
- Non-equiautomic HEA

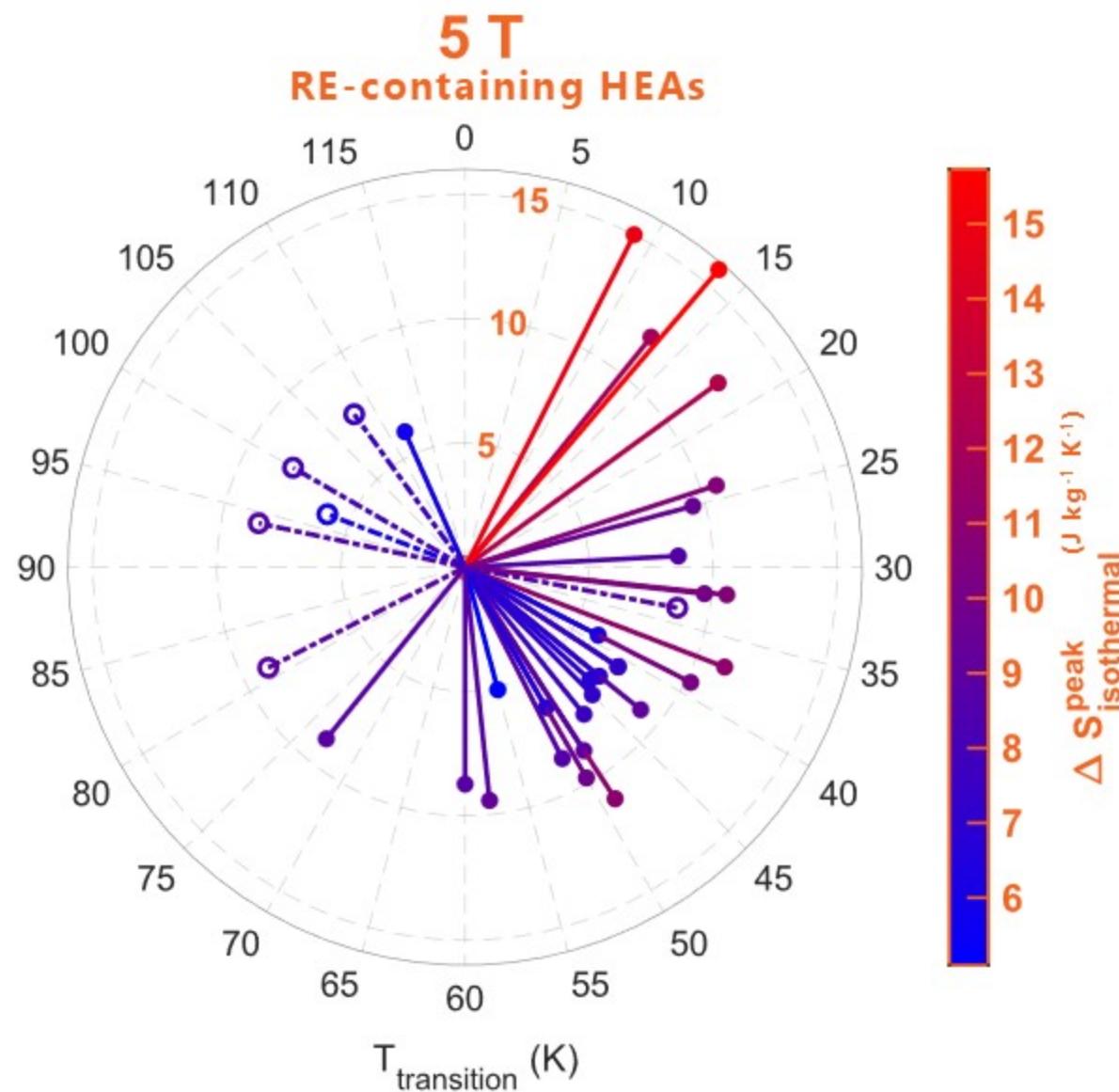
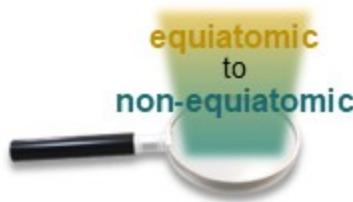
Search strategy

through the large HEA space



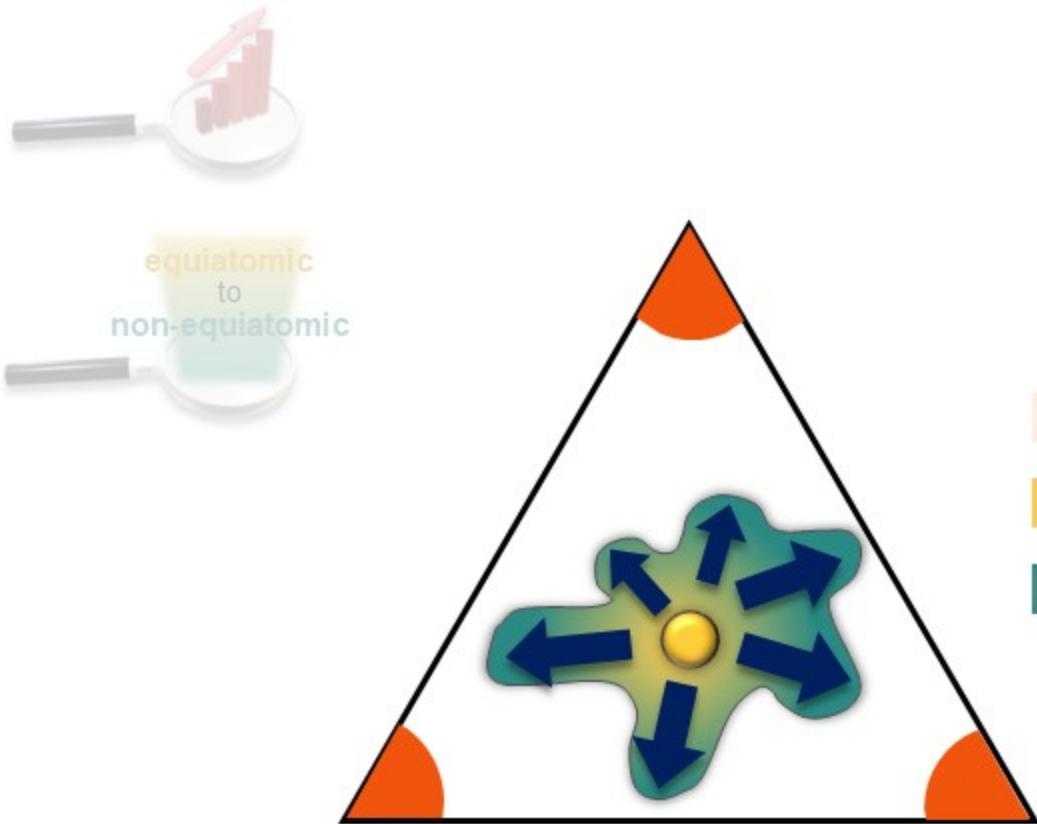
Search strategy

through the large HEA space

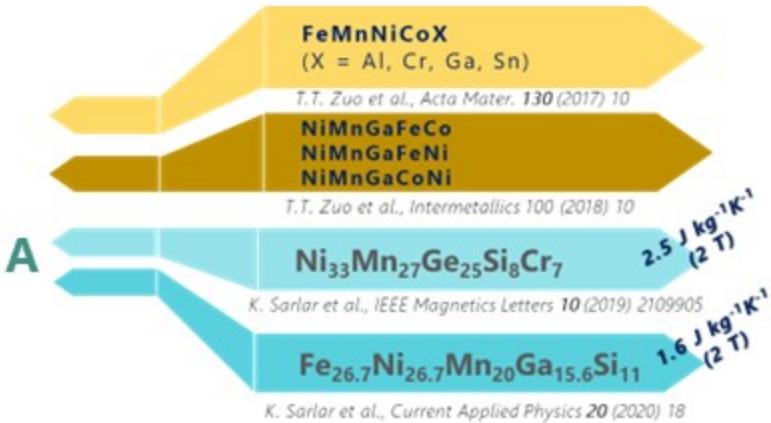


Search strategy

through the large HEA space

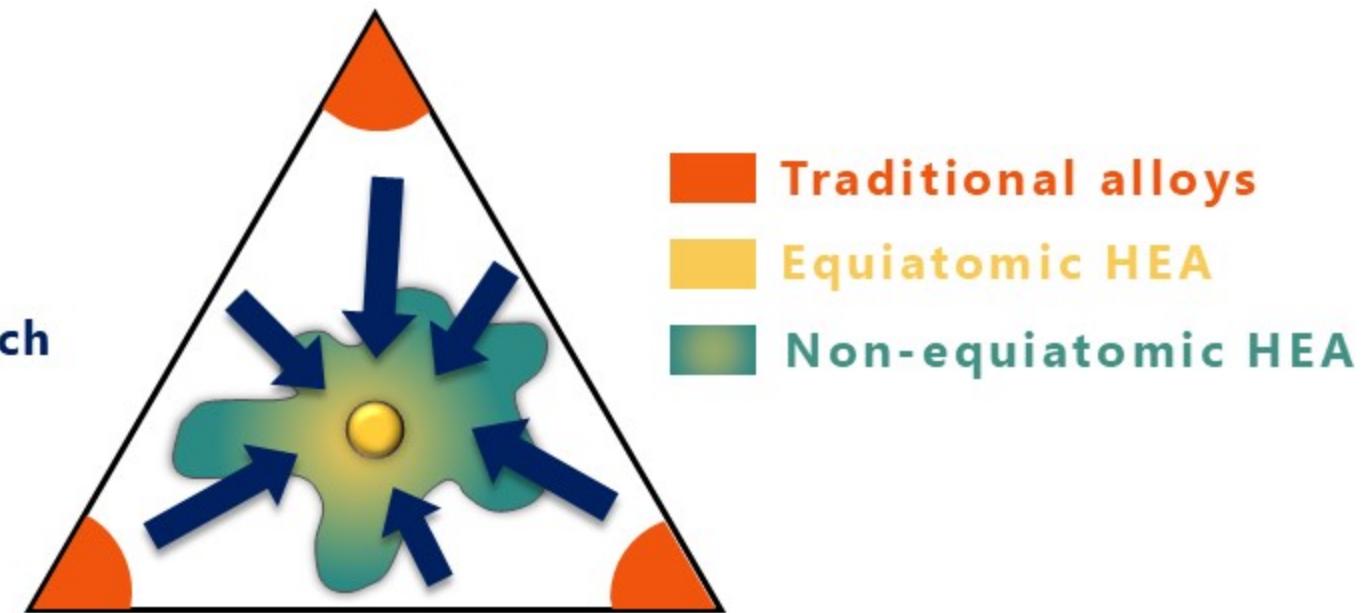
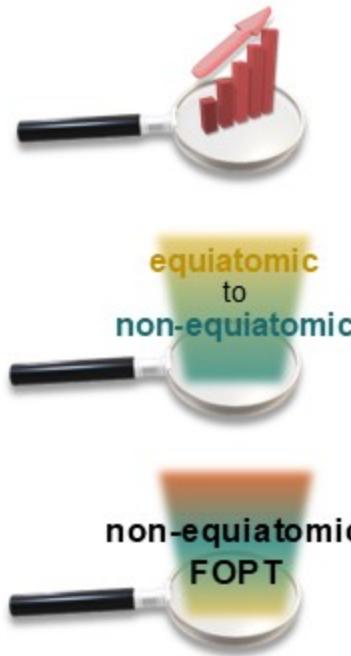


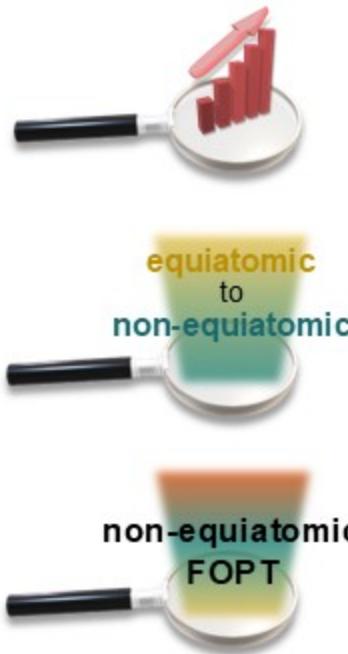
Traditional alloys
Equiatomic HEA
Non-equiautomic HEA
SOPT



Search strategy

through the large HEA space





targeted property search

Frontiers in high entropy alloys and high entropy functional materials

Review | Published: 28 August 2024

Volume 43, pages 4639–4776, (2024) [Cite this article](#)



Rare Metals

Aims and scope →

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3 High-entropy alloys as cutting-edge functional materials

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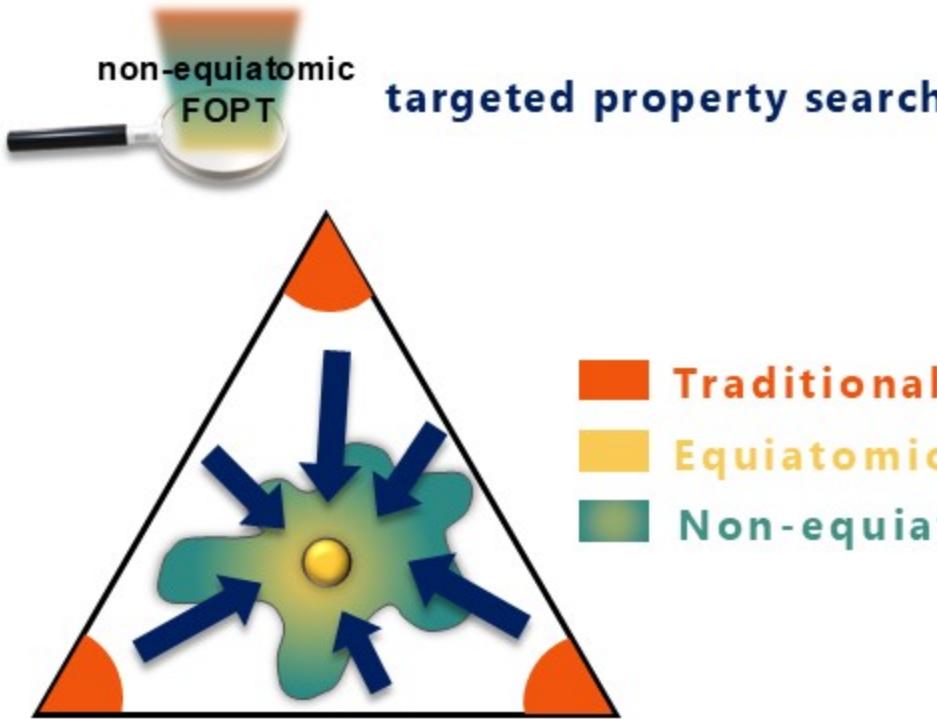
irrev

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 $7.3 \text{ J kg}^{-1} \text{ K}^{-1}$ in $\Delta \mu_0 H = 2.5 \text{ T}$ in $\text{Mn}_{22.3}\text{Fe}_{22.2}\text{Ni}_{22.2}\text{Ge}_{16.65}\text{Si}_{16.65}$ HEA, which was further increased to $13 \text{ J kg}^{-1} \text{ K}^{-1}$ in $\Delta \mu_0 H = 2.5 \text{ T}$ by compositional optimization in the HEA space [629]. Recently, Zheng et al. [630] also reported a large ΔS_{iso} of $48.5 \text{ J kg}^{-1} \text{ K}^{-1}$ under $\Delta \mu_0 H = 5 \text{ T}$ in $(\text{MnNi})_{0.6}\text{Si}_{0.62}(\text{FeCo})_{0.4}\text{Ge}_{0.38}$ 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 $\text{Mn}_{1.75}\text{Fe}_{0.25}\text{CoNiGe}_{1.6}\text{Si}_{0.4}$ 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



Starting composition
selection based on known alloy
with desired behavior



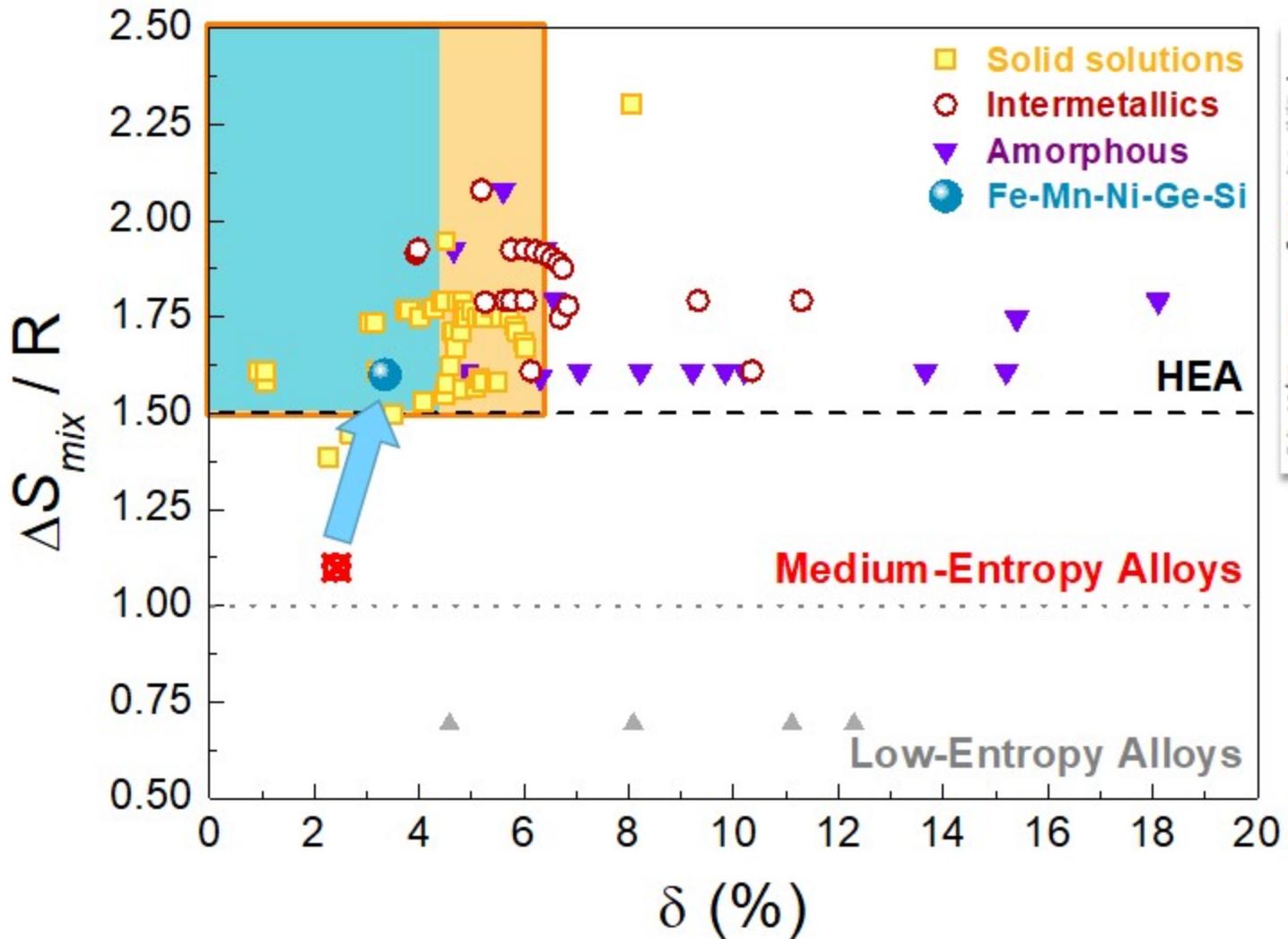
Tune towards HEA region
considering elements with
chemical compatibility



**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

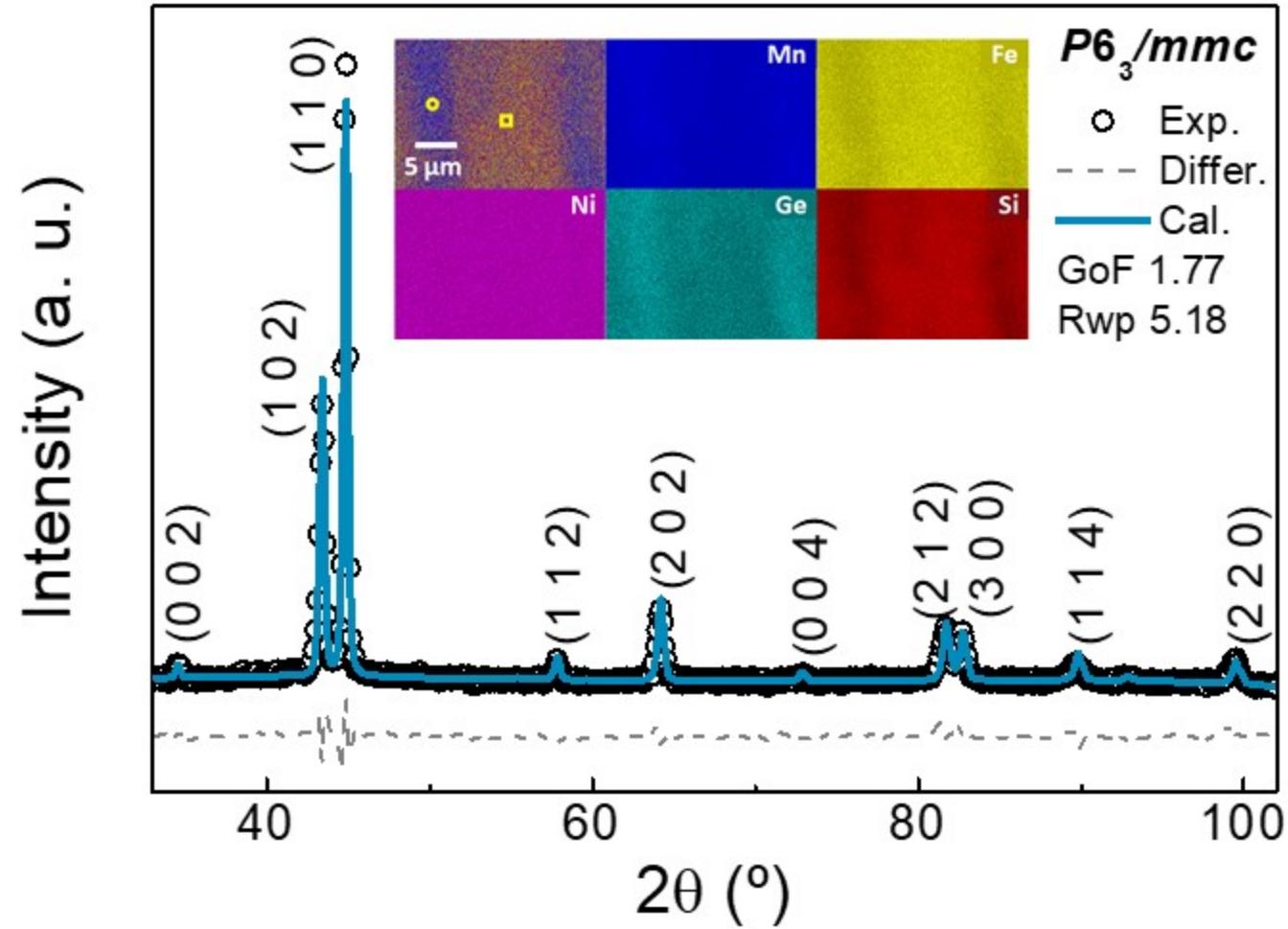
Jia Yan Law ^{a, **}, Luis M. Moreno-Ramírez ^a, Álvaro Díaz-García ^a, Andrés Martín-Cid ^b, Shintaro Kobayashi ^b, Shogo Kawaguchi ^b, Tetsuya Nakamura ^b, Victorino Franco ^{a,*}

^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

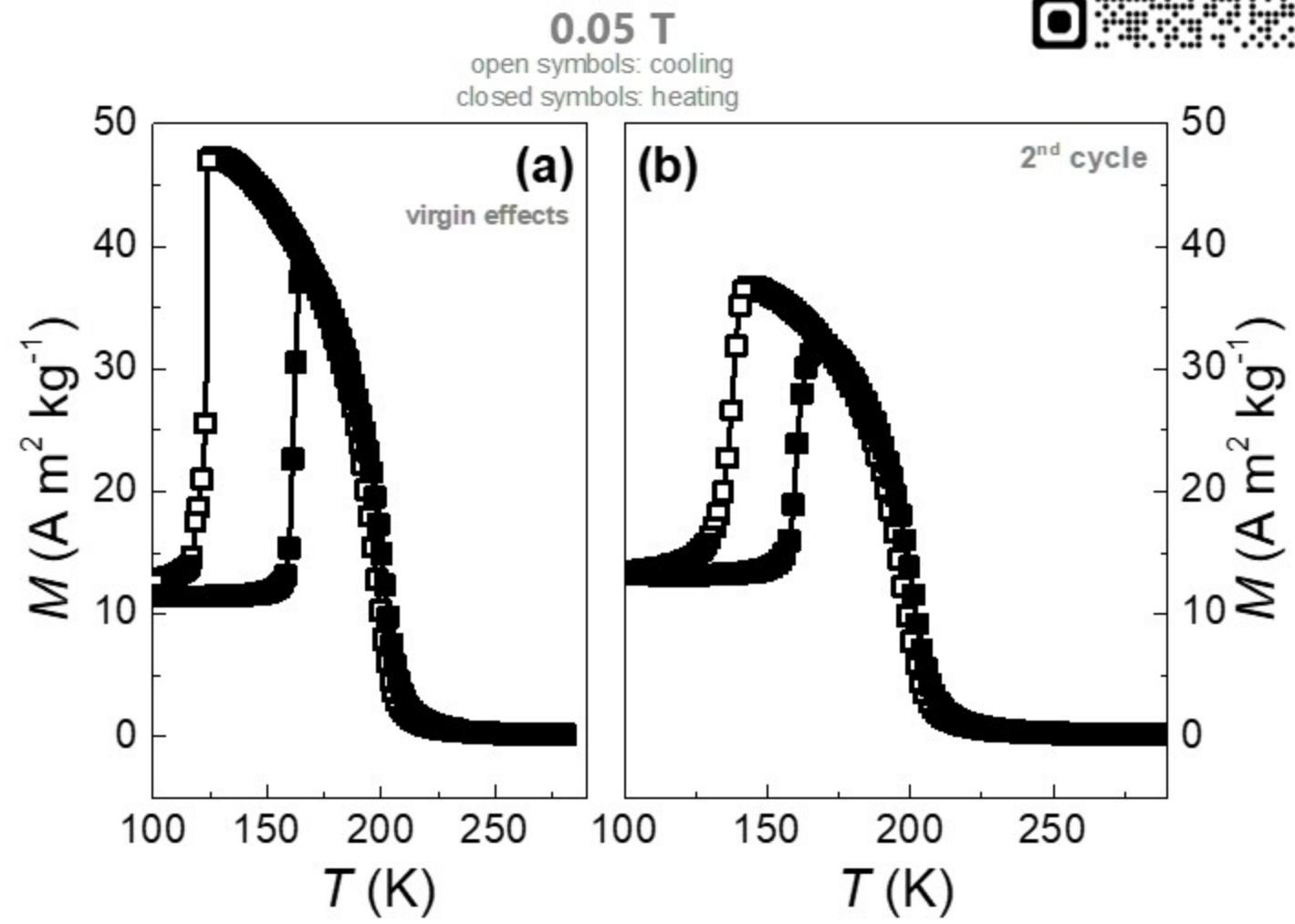
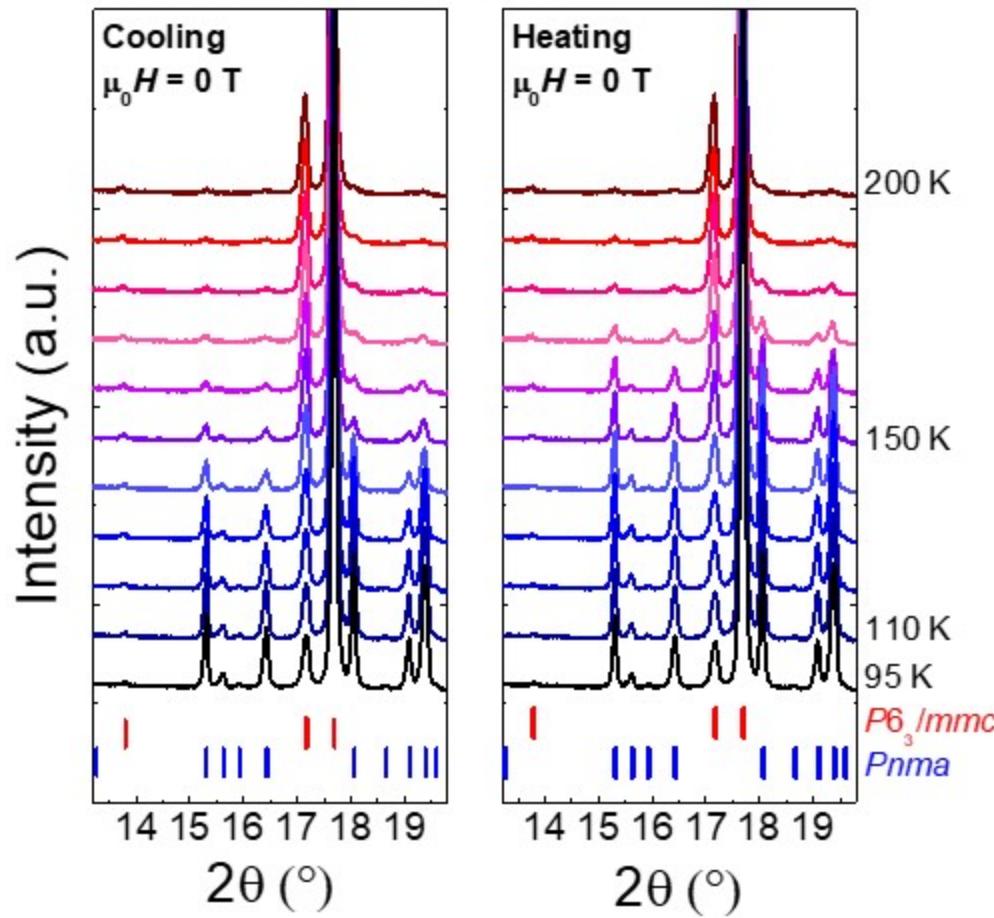
At room temperature



Upon heating and cooling



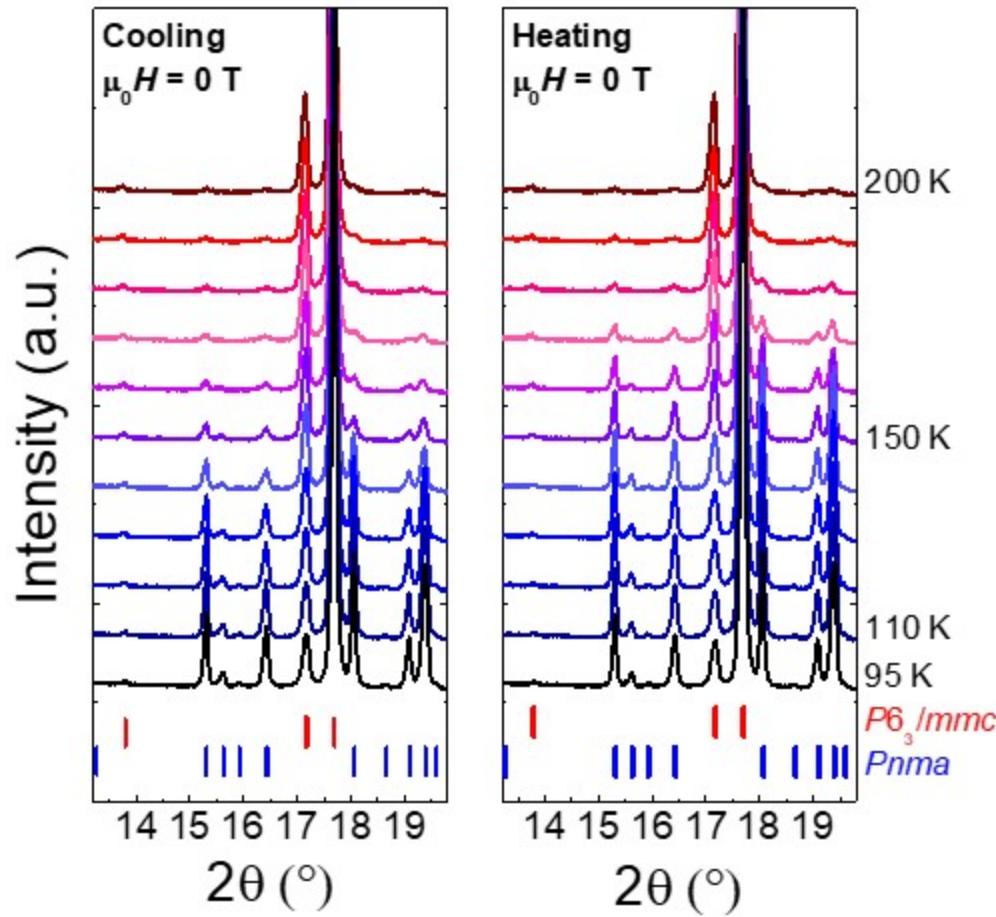
JY Law, L.M. Moreno-Ramírez, Á. Díaz-García, A. Martín-Cid,
S. Kobayashi, S. Kawaguchi, T. Nakamura and V. Franco,
J Alloys Compds 855 (2021) 157424



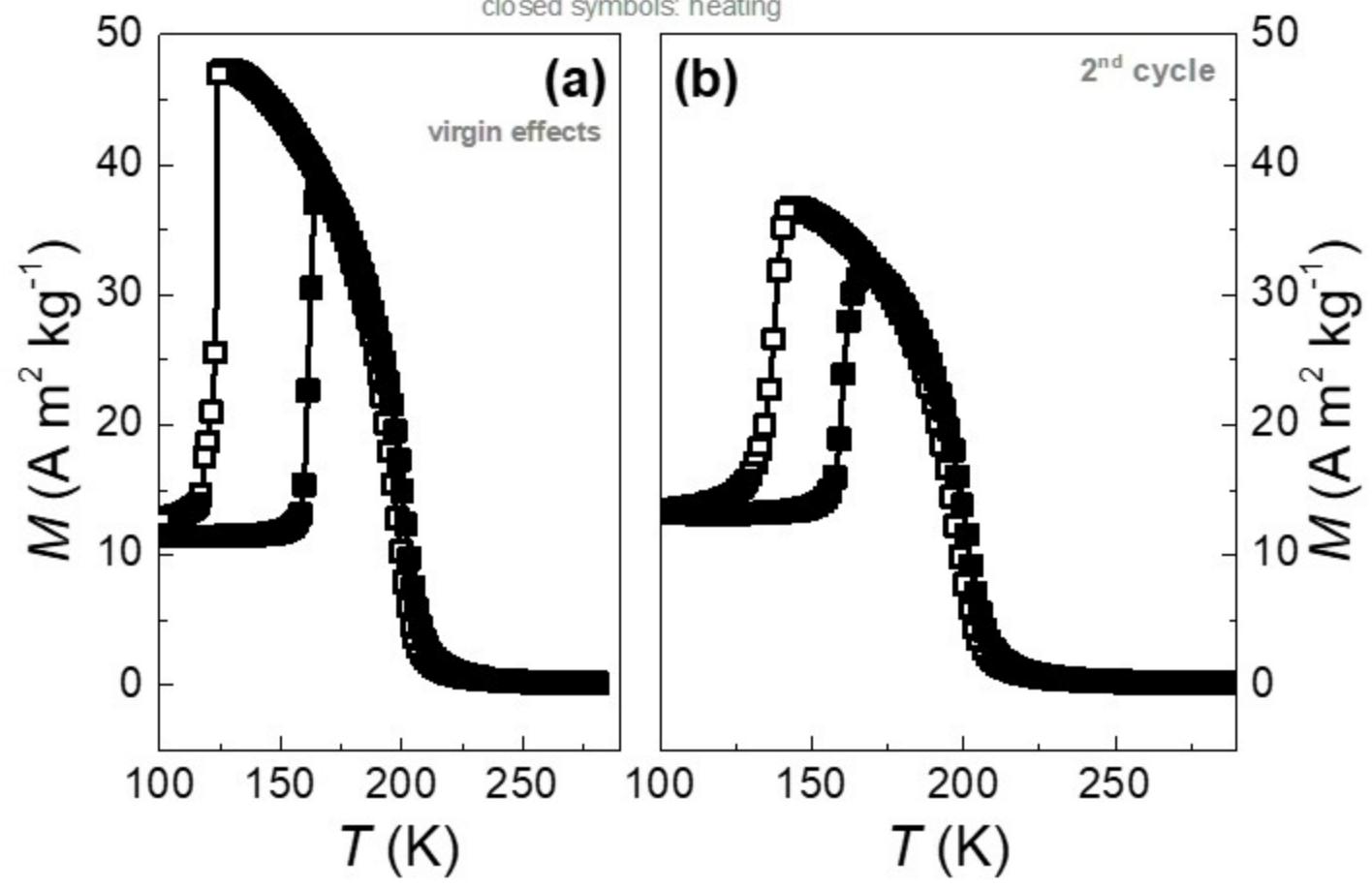
Phase transition temperatures in good agreement

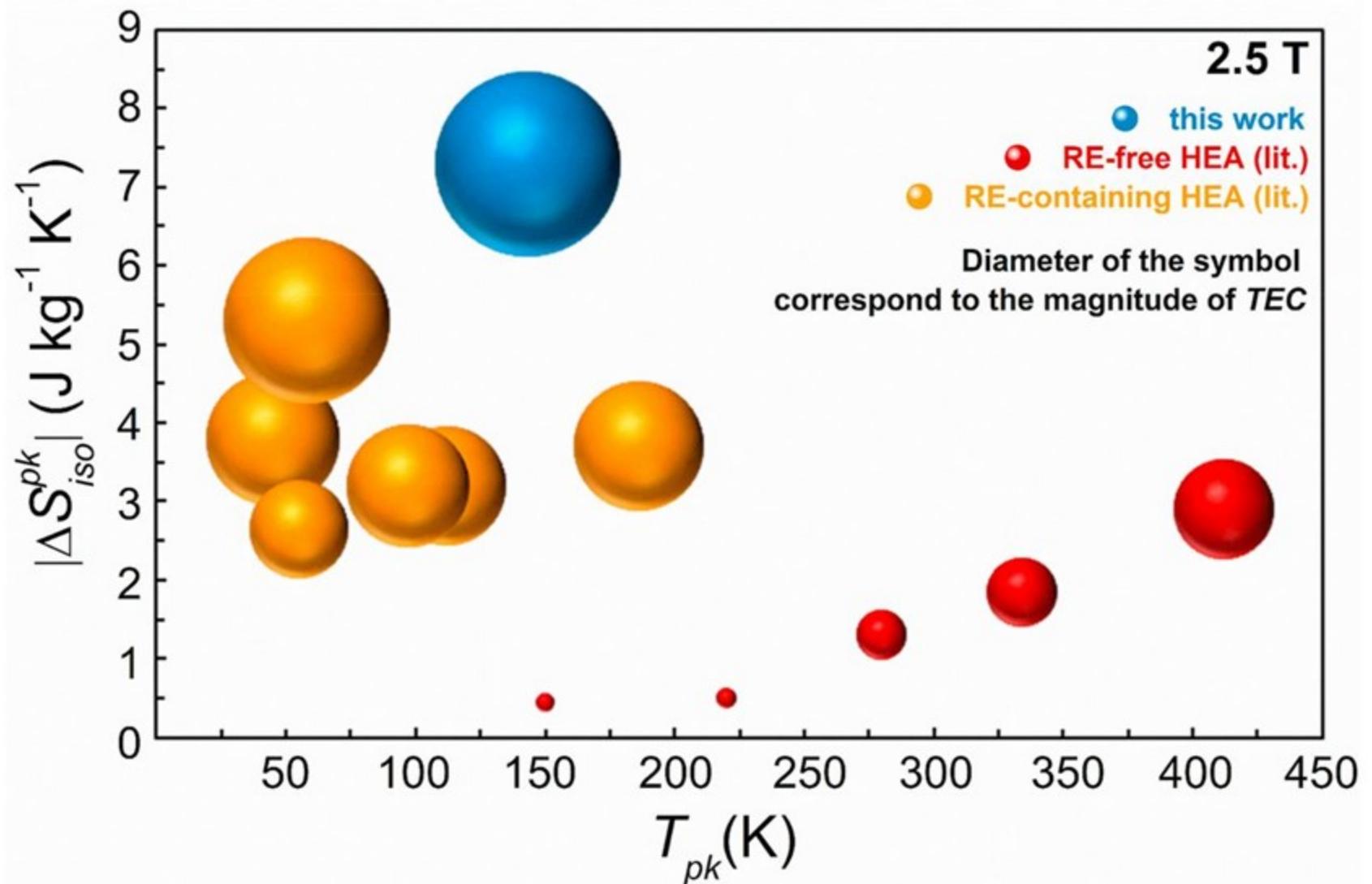
magnetostuctural

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



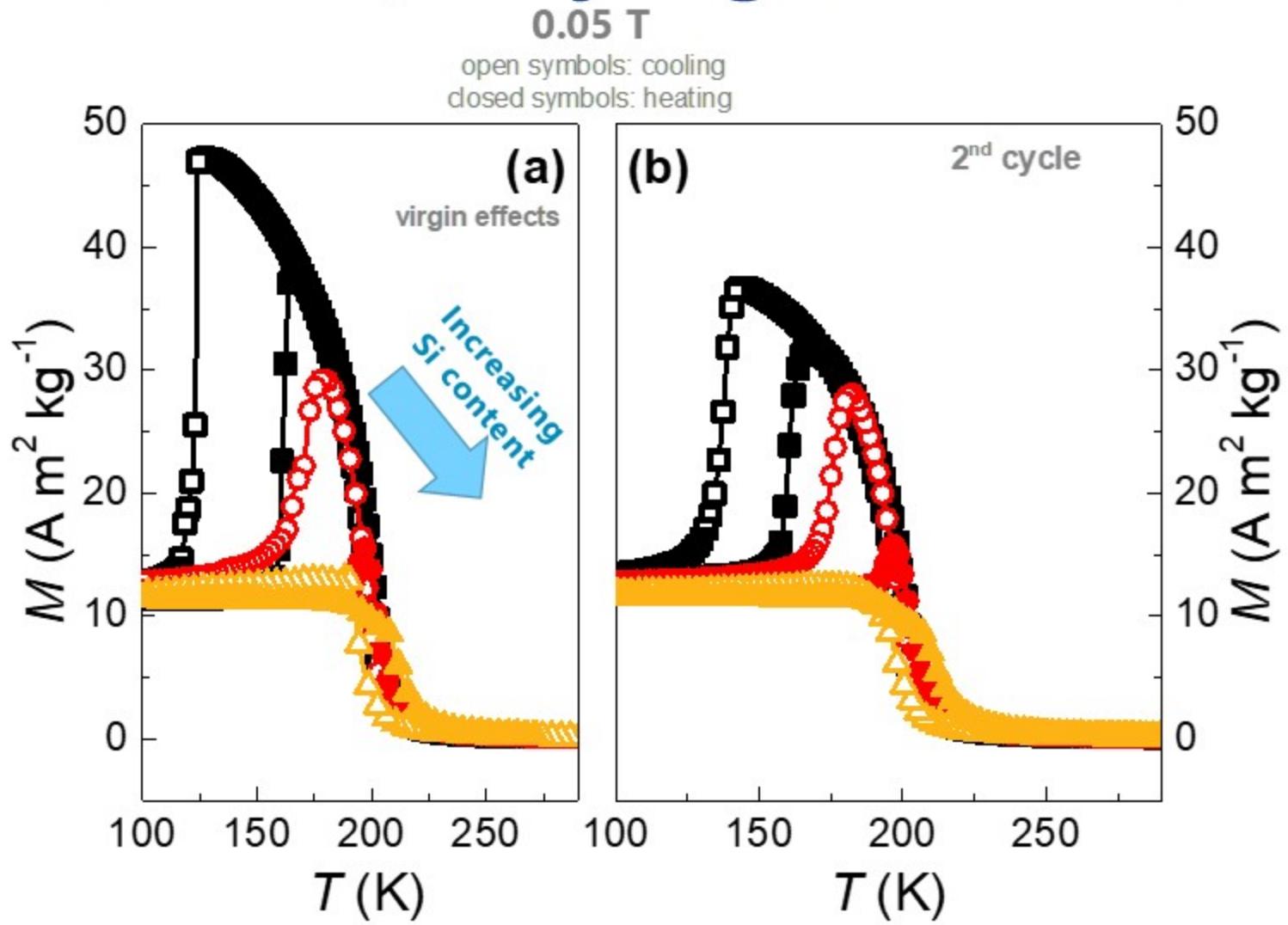
0.05 T
open symbols: cooling
closed symbols: heating





Could that go further?

series (varying Si content)



Acta Materialia 212 (2021) 116031



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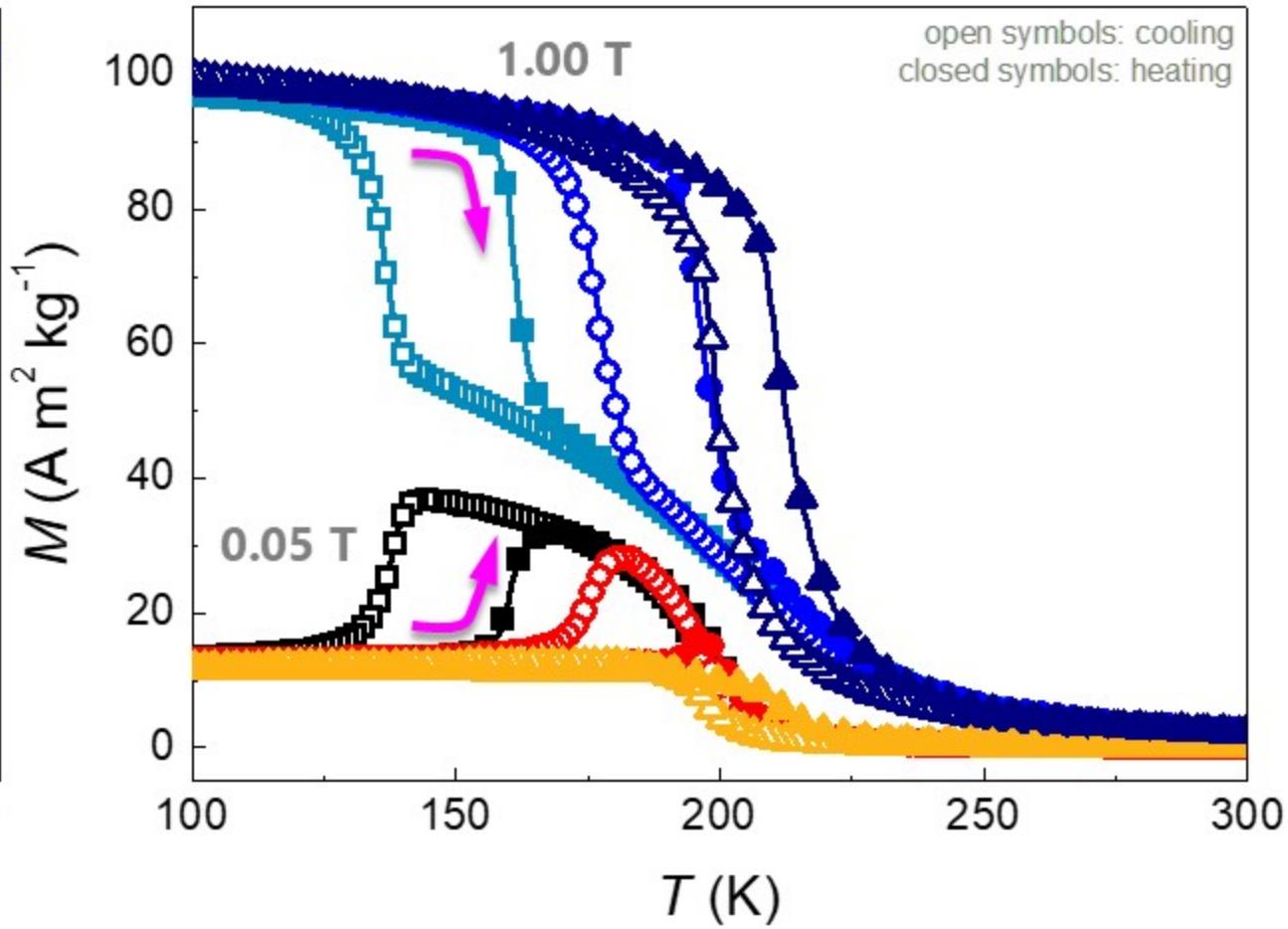
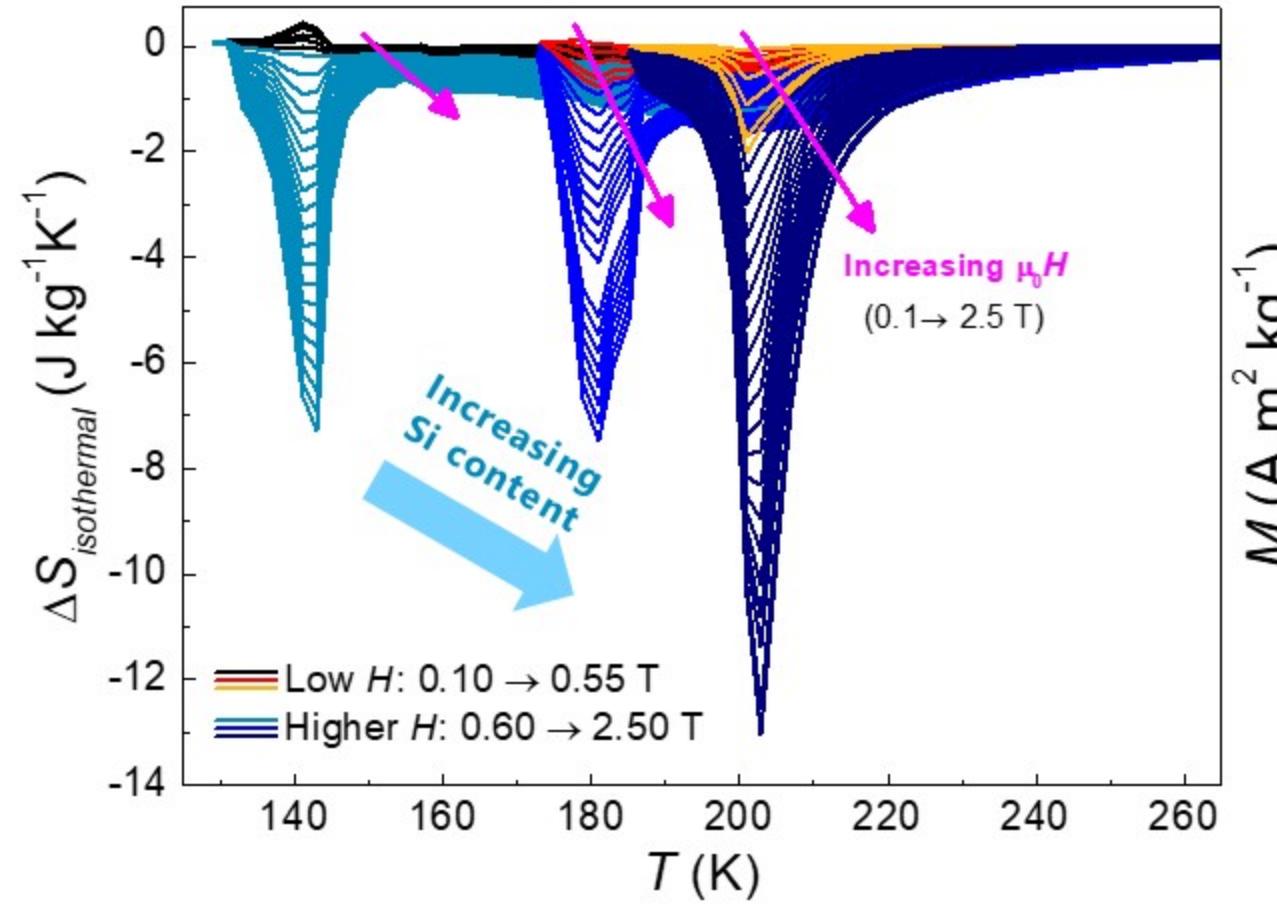
Increased magnetocaloric response of FeMnNiGeSi high-entropy alloys

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

Departamento de Física de la Materia Condensada, ICM-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



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 Law¹, Victorino Franco¹, Luis Miguel Moreno-Ramírez¹, Alejandro Conde¹, Dmitriy Y. Karpenkov², Iliya Radulov², Konstantin P. Skokov² & Oliver Gutfleisch²



order of phase transition

can be determined by exponent n

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

where n depends on temperature and field

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

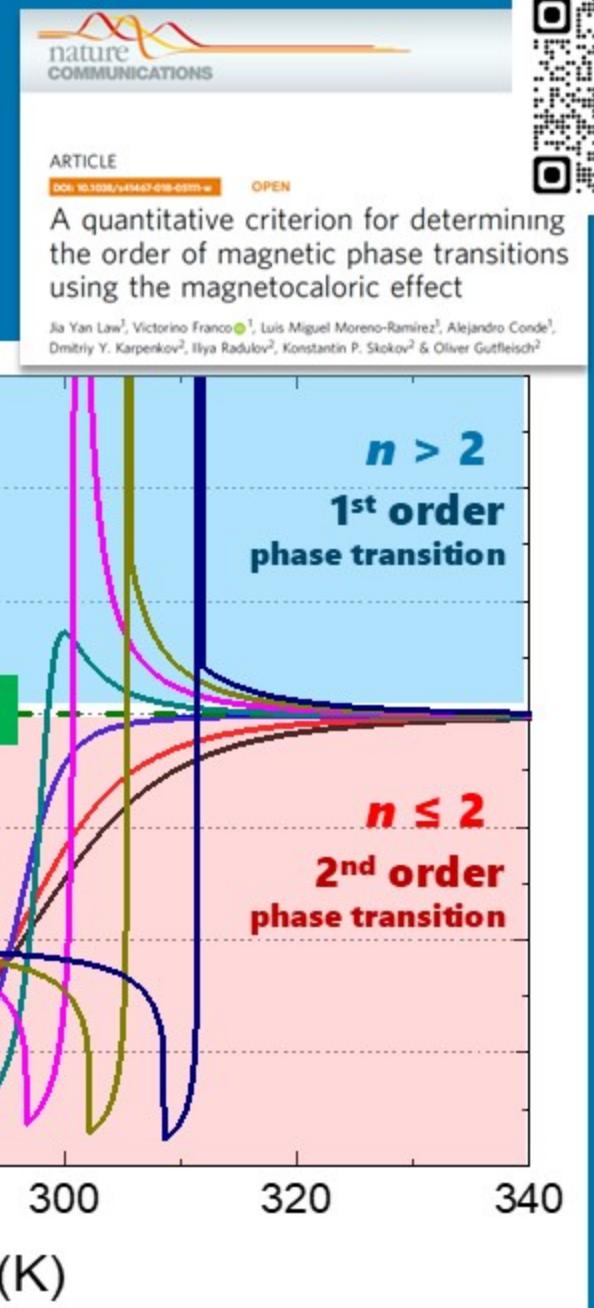
Bean and Rodbell model (order of phase transitions)

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

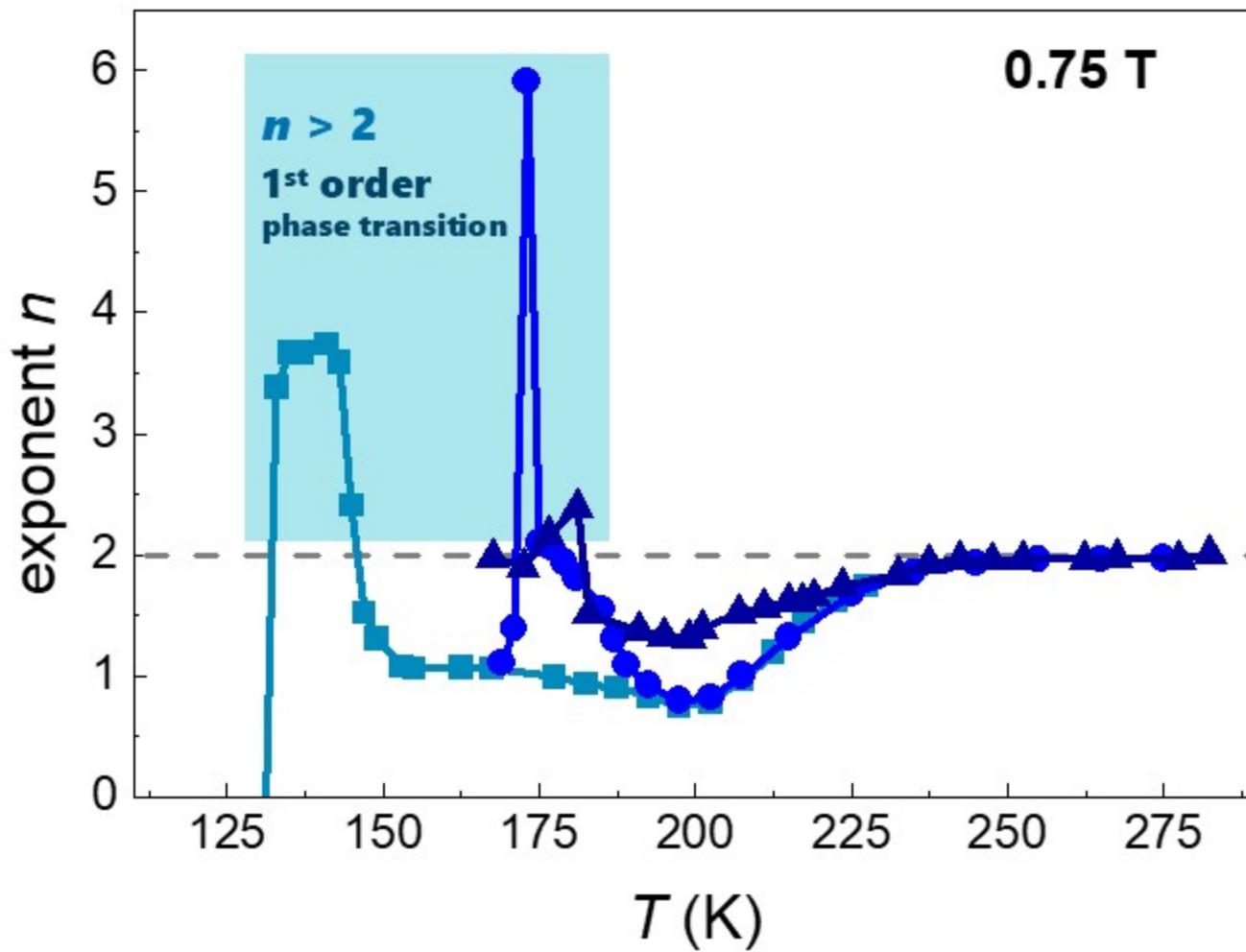
for $\eta = 1$: critical point

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

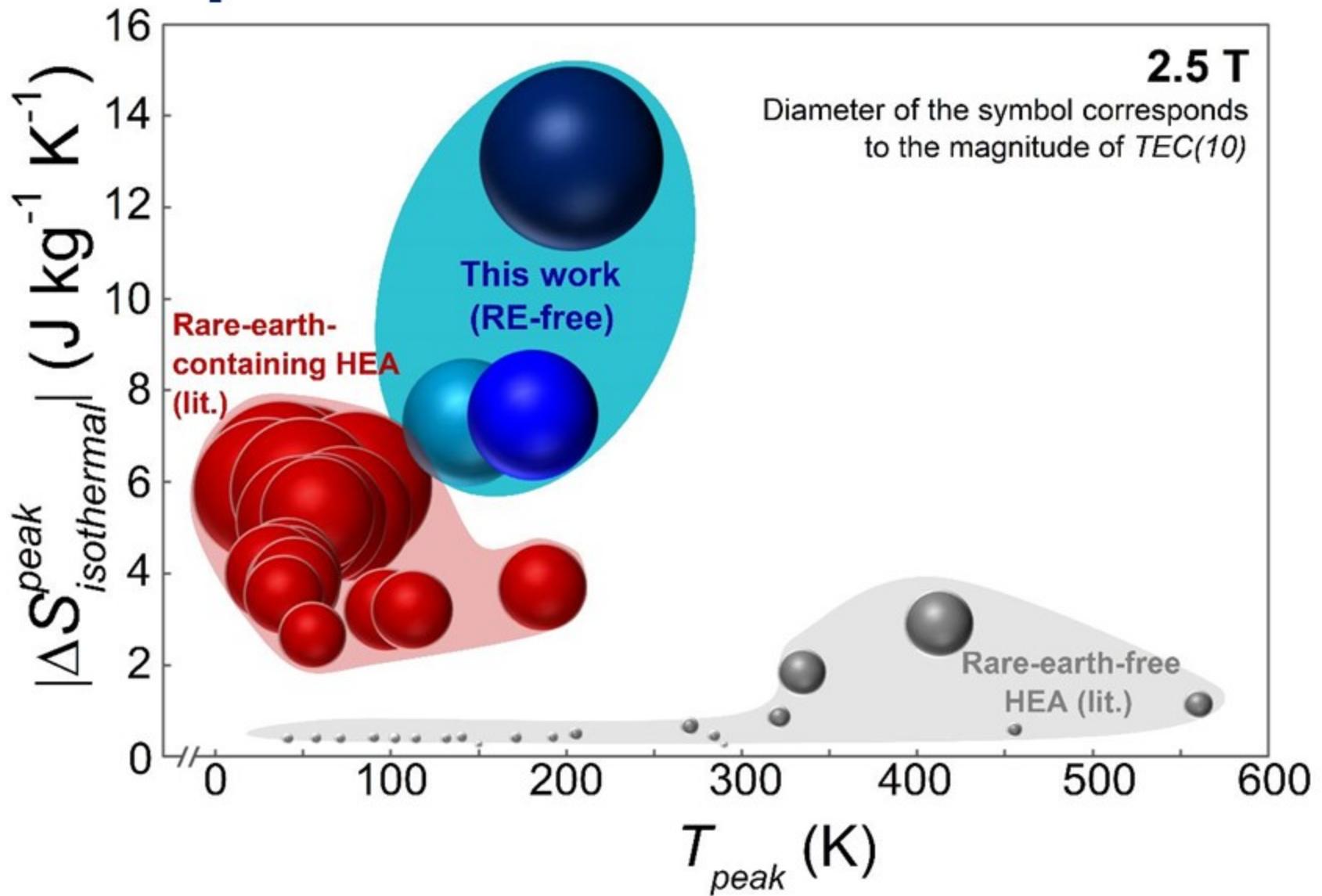
JY Law et al.,
Nature Communications 9 (2018) 2680



order of phase transition

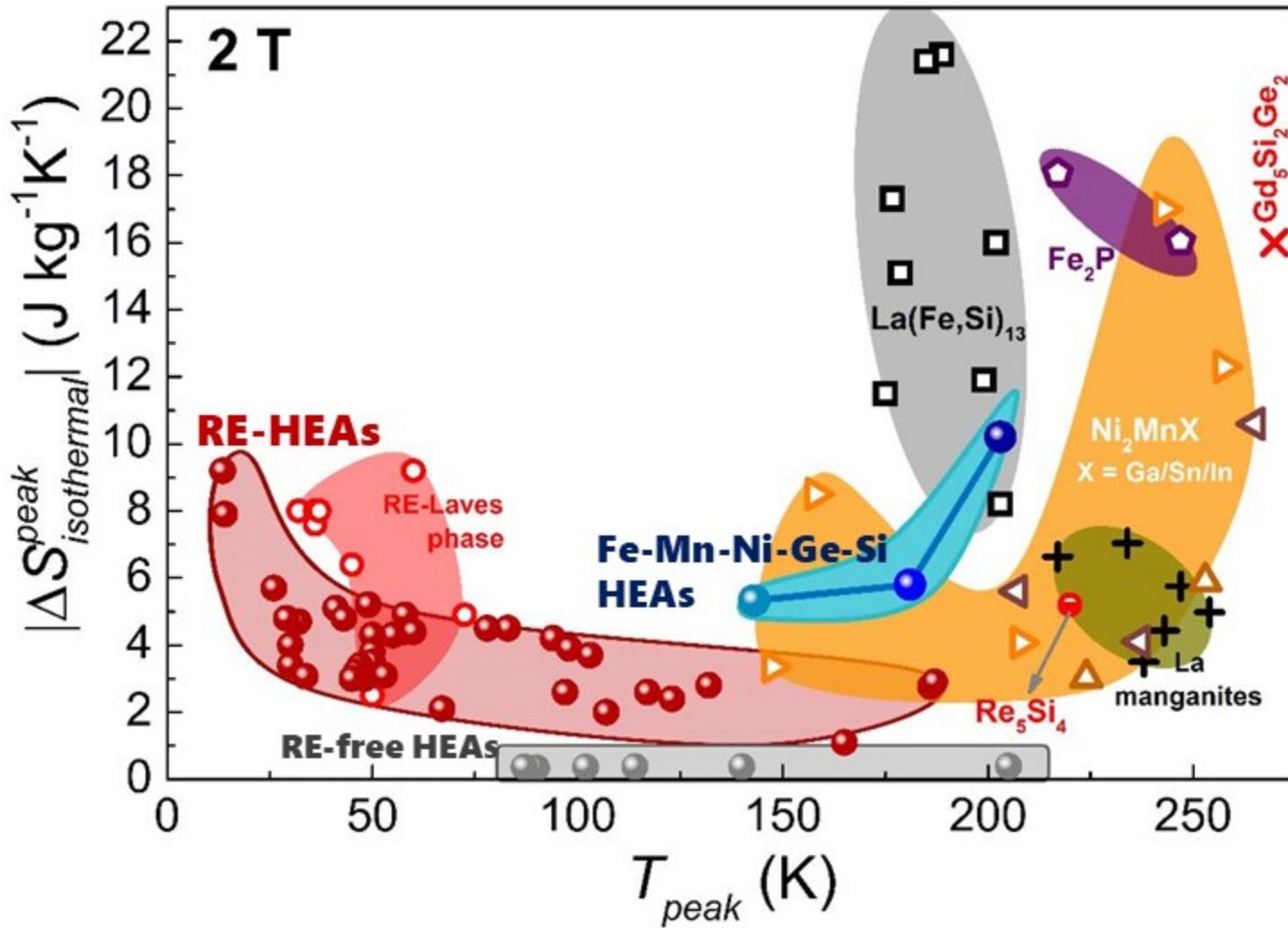


How it compares with literature HEAs



Fe-Mn-Ni-Ge-Si

and what about with conventional MCE materials

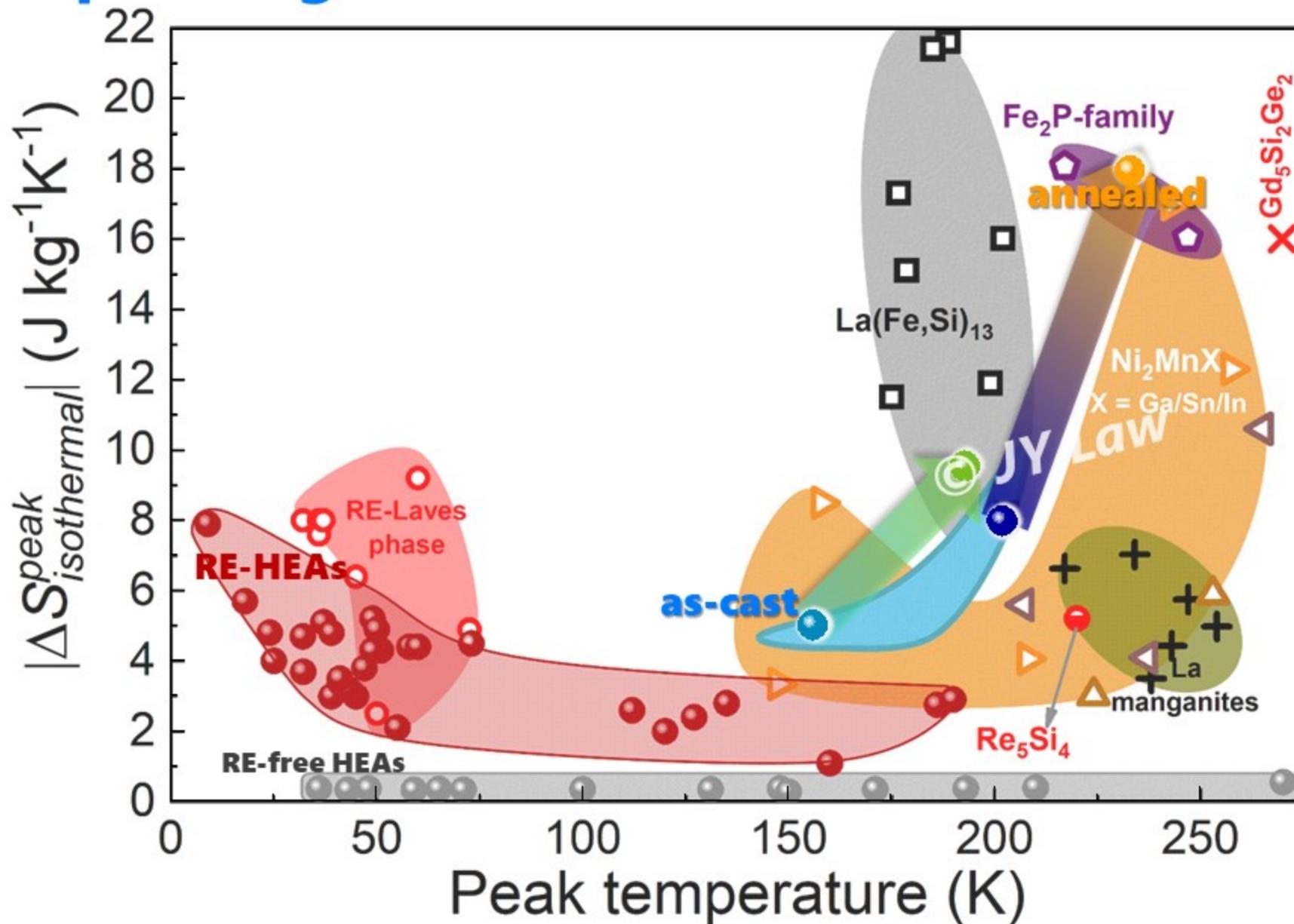


Fe-Mn-Ni-Ge-Si



**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

Acknowledgment

CEMAG

Club Español de Magnetismo



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