



Heusler alloys are a promising class of effective functional materials to be used for green energy technologies. They demonstrate strong coupling between lattice, electronic and magnetic degrees of freedom that allows for a flexible crystallographic and electronic structure.

Disorder in Heusler alloys

Efficient energy conversion by deliberate defects

By Olga N. Miroshkina & Markus E. Gruner

Energy efficient technologies

In the modern world, we consume energy with almost no break: when we use our computers, mobile phones, transport, when we heat our homes, cook food, and make coffee. Although one may not remember it in daily life, an industrial consumption should be taken into account since it is essential for producing all the amenities described above. According to the International Energy Agency (www.iea.org), the total energy consumption has doubled in the last 50 years. Most of this energy is produced with fossil fuels, which include coal, petroleum, natural gas, oil shales, bitumens, tar sands, and heavy oils, and affects the environment negatively. Climate

change, ozone depletion, loss of soil from erosion, biodiversity loss, air and water pollution: We are already entering the phase where we can feel the knock-on effects. This may lead to unpredictable consequences for humanity.

But how can we avoid these devastating consequences and save the environment for our future? The straightforward way is to abstain from using fossil fuels and move towards sustainable, renewable, or alternative energy (so-called “green energy”). However, to be successful on this path, one has to develop efficient functional materials which can be used as a working body in green energy technologies.

Much effort is focused on finding such materials among *crystal-*

line solids. First, let us remember that the solid state is distinguished from the gas and liquid states by a rigid structure in which the constituents (atoms, ions, or molecules) are usually strictly assigned to their positions. In *crystal* or *crystalline solids*, atoms are arranged in a highly ordered microscopic structure, forming a crystal lattice that extends in all directions.

Ferromagnetic materials are of special interest since they are essential components in motors, generators, transformers, and actuators. For example, using *soft* (easy to magnetize and demagnetize) and *hard* (requires a larger magnetic field to become magnetized) magnetic materials, the conversion of electrical energy into mechanical work is per-

formed by electric motors or – for the reverse conversion – generators. Soft magnetic materials are of interest also for inductors/transformers in high frequency power electronics components and power conditioning systems. Due to their large magnetocrystalline anisotropy, *permanent magnets* are vital hard magnetic materials for the replacement of oil-based fuels in transportation by electric motors. *Shape memory* magnetic materials can change their shape under the magnetic field and return to their initial state when the field is removed. These materials can be used for actuators and sensors.

Magnetic materials for new cooling technologies are another important target. A large fraction of electricity consumption is related to cooling systems. The prevailing gas-compressor refrigeration technology was patented back in the middle of the 19th century and has not undergone any fundamental changes since then. Magnetic cooling is an efficient and effective alternative to the gas-compressor cycle. Magnetic refrigeration is based on the intrinsic heating or cooling of a magnetic

material when applying or removing a magnetic field, which is called the magnetocaloric effect. For this technology, high-performing *magnetocaloric* materials need to be developed.

The central question now is: what are the primary requirements for an efficient material? First, an effective magnetic material should consist of the abundant non-toxic elements. Second, it should demonstrate a large desirable effect (magnetocaloric effect, shape-memory effect, large magnetocrystalline anisotropy, etc.). Third, this effect should be obtained in an appropriate temperature range (in most cases, room temperatures). Fourth, there should be no degradation of the material and the effect it demonstrates after many cycles of external load application (magnetic or electrical field, pressure, etc.).

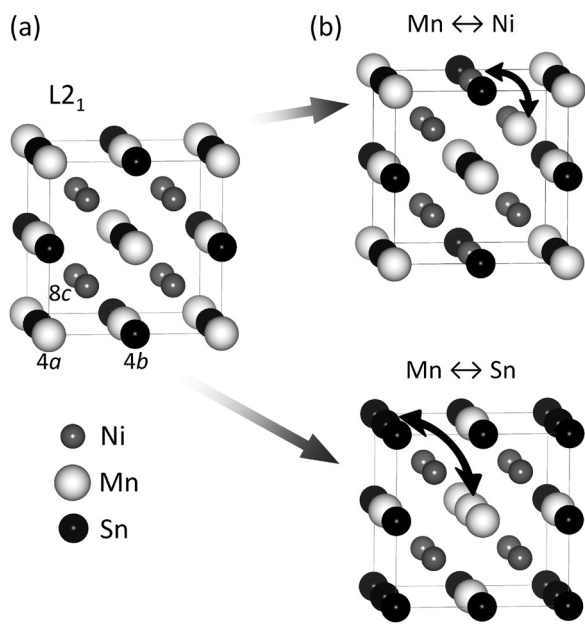
Taking these prerequisites into account, searching for efficient functional magnetic materials is a challenging task for material scientists. Nowadays, there are several classes of the effective magnetic solid-state materials for green energy technologies. One can note $Gd_3(Si_{1-x}Ge_x)_4$,

$La(Fe, Si)_{13}$, Fe_2P -based compounds for magnetocalorics and Fe-Ni, Sm-Co and Nd-Fe-B for permanent magnet application. In this work, we would like to discuss Heusler alloys [1], which are another high-performance material class, and consider their prospects for efficient energy conversion.

Heusler alloys

Heusler alloys are ordered intermetallics with a generic formula (*stoichiometry*) X_2YZ , where X and Y are transition metals (d -block in the periodic system), and Z is the main-group element (p -block). They are named after German mining engineer and chemist Friedrich Heusler, who described the Cu_2MnAl compound in 1903, which is ferromagnetic, despite consisting of non-ferromagnetic elements. According to their chemical formula, the proportion of elements in the full-Heusler alloys is 2:1:1. Looking at the periodic table, one can see that many elements can be combined in Heusler alloys, and a large variety of compositions is possible. Different combinations lead to different magnetic and mechanical properties and functional characteristics, which may or may not meet the application requirements. That is why materials scientists conduct theoretical and experimental investigations of the most promising compositions to advance them further to application.

In addition to a certain formula, X_2YZ Heusler alloys also have a specific structure. In *Strukturbericht* notation, it is referred to as the $L2_1$ structure, and each of the four constituent atoms (as per the formula, these are two X , one Y , and one Z atom) takes a certain position there, as depicted in Fig. 1a, using Ni_2MnSn as an example. In crystallography, these positions are called *Wyckoff positions*, then fractional coordinates are indicated in the brackets. X atoms are located at equivalent $8c$ ($1/4, 1/4, 1/4$) and ($3/4, 3/4, 3/4$) Wyckoff positions, while Y



(1) (a) $L2_1$ Heusler structure, using Ni_2MnSn as an example. Atoms are located in the following positions: $8c$ ($1/4, 1/4, 1/4$) and ($3/4, 3/4, 3/4$) for Ni, $4a$ ($0, 0, 0$) for Mn, and $4b$ ($1/2, 1/2, 1/2$) for Sn. (b) Structures with anti-site disorder introduced by swapping between Mn ↔ Ni and Mn ↔ Sn.

Source: own illustration

and Z atoms occupy the 4a (0, 0, 0) and 4b (1/2, 1/2, 1/2) sites, respectively. The atoms in these positions form *sublattices*. The $L2_1$ structure has a cubic shape and is called the *austenitic* or *parent phase* of an alloy. The sides of this cube all have equal lengths, i.e. $a = b = c$, with the actual length depending on the composition. Under an external load (field, pressure, or temperature), the structure can be compressed or stretched into the so-called tetragonal phase with $a = b \neq c$, thus taking the form of a cuboid. Such structural transitions from the highly symmetric cubic (*austenite*) to the lower-symmetry tetragonal (*martensite*) structure are called *martensitic transitions*. The characteristic feature of this transition is that it is *diffusionless*, i.e. atoms preserve their relative arrangement and the distances of their displacement is smaller than the spacing between them. Heusler alloys undergoing such a transition are of special interest as a martensitic transformation is in many cases the basis for large functional effects.

Besides structural transition, Heusler alloys can also demonstrate transition between different magnetic states. These include *ferromagnetic (FM)*, *ferrimagnetic (FiM)*, *antiferromagnetic (AFM)*, and *paramagnetic (PM)* states. They are the result of different arrangements of the magnetic moments ensuing from the intrinsic magnetism of the electrons of each element with respect to one another. *Magnetic moment* can be described as a vector quantity, for which both magnitude and direction (orientation) are important. The total magnetization is defined as the quantity of magnetic moment per unit volume, and it characterizes the magnetic properties of the entire compound, in terms of the sum of the individual constituents (atoms). If the magnetic moments of all atoms are oriented parallel to one another, the material is in the FM state, and the total magnetization of the compound is maximized. If some magnetic moments are ori-

ented antiparallel to the others (in notation, some would be pointing up and some down) and the numbers of ‘ups’ and ‘downs’ and/or their magnitudes are different, this results in an uncompensated FiM configuration. The total magnetization of the FiM phase is thus not zero but smaller than the FM. When half of the atoms have magnetic moments oriented up and the other half is oriented down, they compensate one another, and the resulting total magnetization is zero. This is called the AFM state. A PM configuration is randomly disordered, i.e. the individual magnetic moments are oriented chaotically, which also results in zero total magnetization. Under certain conditions (temperature, magnetic field), a compound can pass from one magnetic state to another. Such a *magnetic transition* can accompany structural transformations or take place independently (at lower or higher temperatures than the martensitic transition temperature) depending on the composition. Thus, we can control the sequence of the magnetic and structural transitions by varying only the proportion of the constituent elements.

This helps us answer the question ‘Why do Heusler alloys attract so much attention?’ The first two reasons are simple. Heusler alloys can be formed without rare and toxic elements, which makes them ecologically friendly. They can also consist of relatively cheap, abundant elements. However, the most important reason is the exceptional tunability of their crystallographic and magnetic structure, which gives rise to many functional properties.

In the following sections, we will show how one can tune Heusler alloys’ properties by introducing different types of disorder. In our minds, ‘disorder’ often has a negative connotation and is associated with something being wrong or improper. But this is not always the case, especially if it is applied in a controlled fashion. In mate-

rial synthesis, one can deliberately introduce disorder and thus change the material’s properties such as the temperature at which structural or magnetic transformation takes place, magnetic response or the magnitude of the effect. In our theoretical investigations, we use so-called *first-principles methods*, which are based on quantum mechanics and statistical physics and do not use any experimental data as input. The only information in such calculations is the atomic number, which determines the number of electrons, the atomic weight of the constituents that form the material and the structure in which they are arranged. Our first-principles method of choice is *density functional theory (DFT)*. The idea behind DFT is to express the total energy of a many-electron system (compound) in terms of the total electron density rather than the complex quantum mechanical wave function. This allows a significant decrease in the computational cost of calculations while yielding reasonable results using appropriate approximations. With the help of DFT, we consider two examples where introduced disorder changes the structural and magnetic properties of Heusler alloys significantly and explain the underlying mechanisms.

Effect of positional disorder on the structural and magnetic properties of Ni_2MnSn

Bearing in mind that Heusler alloys are supposed to have an ordered $L2_1$ crystal structure, we modelled the ideal $L2_1$ structure for FM Ni_2MnSn to analyze its properties. One way to determine the thermodynamic properties of a structure is to look at its *vibrational density of states (VDOS)*. Atoms in a crystal lattice are not ‘frozen’ in their sites but oscillate near their equilibrium position with a small amplitude. Since the lattice is rigid, the displacement of one or more atoms from equilibrium gives rise to a set of waves,

which may propagate throughout the lattice. The VDOS provides a statistical measure of these vibrations and serves as a fingerprint of the interatomic forces, i.e. it shows how atoms affect each other. A *phonon* is the quantized excitation of such a wave in a crystal lattice that arises from oscillating atoms. Every atom contributes to the lattice vibrations, and we can capture all contributions with the help of DFT. We had the opportunity to compare our calculations with an experiment carried out by Prof. Dr. Heiko Wende's research group using nuclear resonant inelastic x-ray scattering (NRIXS), which allowed us to gather information about the vibrations of Sn atoms. Therefore, we will further focus on Sn VDOS to consider the same issue from both an experimental and a theoretical perspective [2].

From the experimental Sn VDOS, the largest Sn contribution is found for low-frequency vibrations (Fig. 2). Meanwhile, the DFT Sn VDOS in the ideal $L2_1$ structure differs qualitatively in this respect and shows a high peak in the high energy range. The overall shape of the low energy parts is reproduced very well, but it also appears to be sharper than the theoretical values. Moreover, ^{119}Sn -Mössbauer measurements, which allow experimental insights into magnetic moments, showed that there are several types of magnetic moments. Usually, in the ordered FM $L2_1$ system, atoms of the same species are expected to have equal magnetic moments (in our case, approximately $0.2 \mu_B$ for Ni, $0.05 \mu_B$ for Sn and $3.6 \mu_B$ for Mn). Thus, we concluded that the reason for the discrepancy between the DFT-calculated and the measured Sn VDOS might be due to a disorder that had not been included in our DFT calculations yet. Disorder leads to changes in the environment of each atom and, as a consequence, changes in the individual magnetic moments and interatomic forces. Even highly ordered experimental samples might still be slightly disordered in terms of their

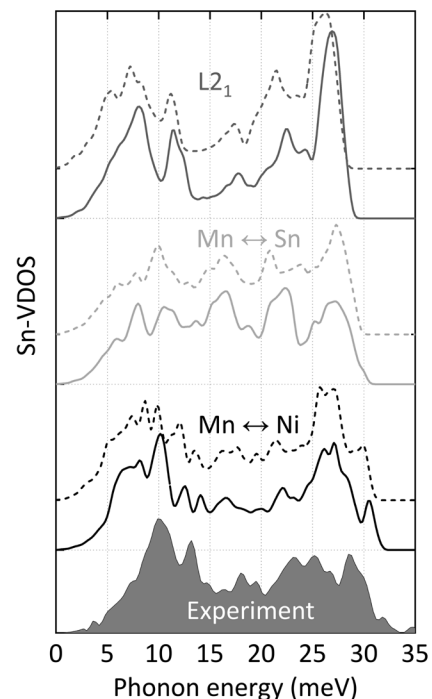
atomic arrangement. To check this, we modelled structures with so-called *anti-site disorder*, which means that some pairs of atoms are swapped compared to the initial $L2_1$ structure. We considered the swapping between $\text{Mn} \leftrightarrow \text{Ni}$ and $\text{Mn} \leftrightarrow \text{Sn}$ as depicted in Fig. 1b. Additionally, for each case, magnetic disorder was introduced through antiparallel orientation of the magnetic moment of one Mn atom (i.e. turning it 'down'). This results in a FiM alignment.

Our calculations show that swapping $\text{Mn} \leftrightarrow \text{Ni}$ and $\text{Mn} \leftrightarrow \text{Sn}$ results in two or three magnetically different types of Sn atoms respectively. In line with the experiment, this suggests a partially disordered sample. For further confirmation, we calculated the VDOS for the disordered structures. Fig. 2 shows that anti-site disorder leads to a characteristic change in the Sn VDOS. First of all, the high-energy contribution is diminished. The FM alignment is in better agreement with the findings from the experiment compared to the FiM arrangement, which also matches the enhancement of the Sn VDOS features at high energies. We do not obtain a perfect fit between the experiment and either case of site disorder, only partial improvements for different parts of the spectra. However, we can expect a real sample to be subject to a complex superposition of different structural and magnetic defects.

This example shows that Sn VDOS is very sensitive to magnetic and chemical disorder at the *d*-metal sites. Sn can 'feel' the changes in its environment even if the Sn atoms themselves were not included in the swapping (as in the $\text{Mn} \leftrightarrow \text{Ni}$ case). Thus, the Sn VDOS can be used as a gauge of the degree of long-range magnetic and structural disorder.

Effect of chemical disorder on the magnetic properties of Ni-Mn-(In,Sn)

When the proportion and/or number of the constituent elements differ



(2) Calculated Sn VDOS for the ideal $L2_1$ and disordered structures of Ni_2MnSn in comparison with the experiment (grey shaded area). Results of DFT calculations are presented for FM (solid lines) and FiM (dashed lines) ordering.

Source: adapted with permission from Ref. [2]. Copyrighted by the American Physical Society

from the generic formula X_2YZ , the composition is called *off-stoichiometric*. In off-stoichiometric compounds, the initial proportion 2:1:1 is modified. Some elements can be supplied in excess and thus occupy the sites of others the supply of which is short, with the total number of atoms per unit cell remaining four. Adding a fourth or even fifth element at the expense of others is also possible. The fourth element must then take the position of one of the initial *X*, *Y* or *Z* atoms, thus decreasing their concentration. The deviation from the stoichiometry usually results in *chemical disorder*. This disorder significantly changes the electronic structure of the system and the interaction between the atoms. When the effects of these changes are understood, one can use the disorder to manipulate the properties of the compound and tune the desirable effects.

We will now illustrate how changing the proportion of p elements in Z positions affects the interaction between other elements and the magnetic transition temperature [3]. We consider a rather complex $\text{Ni}_{1.92}\text{Mn}_{1.44}\text{In}_{0.64-x}\text{Sn}_x$ ($0 \leq x \leq 0.64$) compound. Here, the sublattice of the Z element (4b) is shared between the In and Sn atoms (see Fig. 3a). Also, the Mn concentration is higher than 1 per unit cell. Thus, there is not enough space for all Mn atoms in their original 4a positions, and the excess Mn atoms take the places of their neighbours. They occupy the sites of missing Ni atoms and the In/Sn 4b sublattice.

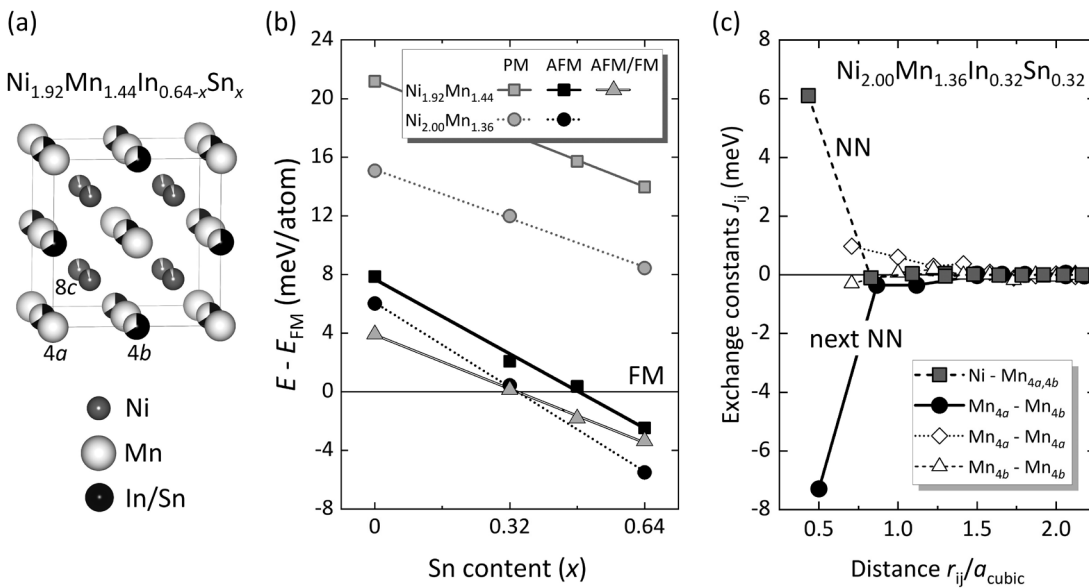
The Curie temperature T_C is the temperature above which FM material becomes PM, i.e. magnetically disordered. Usually, the magnetic moments of all sublattices in Heusler alloys become randomly oriented above the Curie point. However, complex atomic arrangements lead to non-trivial behaviour. Our colleagues from the University of Parma found an effective magnetic decoupling between 4a and 4b sub-

lattices in $\text{Ni}_{1.92}\text{Mn}_{1.44}\text{In}_{0.64-x}\text{Sn}_x$ in the course of an experiment. This means that the sublattices become disordered at different temperatures, i.e. they have different magnetic transition temperatures. On the 4a sublattice, the Mn atoms retain their FM order up to T_C , which changes only slightly with varying compositions. Meanwhile, the FM order on the 4b sublattice weakens and breaks down prematurely with increasing Sn content. Our DFT calculations revealed the mechanism behind this counterintuitive phenomenon.

We started out with $\text{Ni}_{2.00}\text{Mn}_{1.36}\text{In}_{0.64-x}\text{Sn}_x$, which has no Mn atoms in the Ni sublattice. Comparing the total energies, we can determine the energetically most favourable magnetic state. Large-scale DFT calculations reveal that there is a composition-dependent competition between the AFM and FM phases as a function of the Sn content. The FM ground state on the In-rich side turns into an AFM state on the Sn-rich side (Fig. 3b). In the case of $\text{Ni}_{1.92}\text{Mn}_{1.44}$ -based compositions, the excess Mn on the Ni

sublattice (Mn_{8c}) always prefers an arrangement that is antiparallel to Mn_{4a} . With opposite orientation, the structure turned out to be unstable or yield prohibitively high energy. We considered a mixed AFM/FM configuration, where the strong effective FM coupling is mediated by the nearest neighbour (NN) Mn_{8c} . In this case, the orientation of Mn_{4b} is AFM if it has only Ni atoms as NN but FM if Mn_{4b} has at least one Mn_{8c} as NN. Our calculations prove that this fosters the formation of a mixed phase with FM clusters in the AFM matrix on the 4b sublattice. With increasing Sn content, the stability of the mixed magnetic state decreases with respect to a pure AFM arrangement (Fig. 3b).

To evaluate the strength of interatomic interactions and reveal the prevailing ones, we calculated the magnitude of the parallel and antiparallel couplings in terms of so-called *exchange interaction constants*. Fig. 3c shows that the magnetic order is dominated by the competition between the direct inter-sublattice NN exchange between Mn and



(3) (a) Chemical disorder in the quaternary $\text{Ni}_{1.92}\text{Mn}_{1.44}\text{In}_{0.64-x}\text{Sn}_x$ Heusler alloy. Excess Mn atoms go to Ni and In/Sn sublattices. (b) Total energies of different magnetic arrangements (PM, AFM, AFM/FM) relative to the FM state of the respective composition (zero line) as a function of the Sn content according to DFT calculations. (c) Exchange constants J_{ij} of $\text{Ni}_{2.00}\text{Mn}_{1.36}\text{In}_{0.64-x}\text{Sn}_x$ as a function of distance r_{ij} between the interacting lattice sites i and j in units of the lattice constant a_{cubic} obtained with an FM reference state. $J_{ij} > 0$ indicate FM interaction, while $J_{ij} < 0$ correspond to AFM exchange.

Source: figure (a), own illustration, figures (b) and (c) adapted with permission from Ref. [3]. Copyrighted by the American Physical Society.

Ni and next-NN exchange between Mn on different sublattices, Mn_{4a} - Mn_{4b} . Intra-sublattice interactions (Mn_{4a} - Mn_{4a} and Mn_{4b} - Mn_{4b}) remain small according to the larger distances between these atoms. The additional *sp* electron provided by Sn strengthens the AFM Mn_{4a} - Mn_{4b} and weakens the FM Ni-Mn coupling. This results in an antiparallel arrangement of 4a and 4b for Sn content $x > 0.32$.

This result demonstrates how disorder in the transition metal site in combination with variation of the main-group element allows us to control magnetic order in different sublattices in off-stoichiometric Ni-Mn-(In,Sn).

Conclusions

Heusler alloys are a promising class of effective functional materials to be used for green energy technologies. They demonstrate strong coupling between lattice, electronic and magnetic degrees of freedom that allows for a flexible crystallographic and electronic structure. In our work, we showed how this flexibility can be used to control particular vibrational and magnetic properties. We demonstrated how the redistribution of the transition metal atoms together with variation of the main-group element enables fine-tuning of Heusler materials' properties, thus increasing the range of their potential applications.

Zusammenfassung

Magnetische Funktionsmaterialien für saubere und energieeffiziente Technologien müssen unterschiedlichste Voraussetzungen erfüllen. Die Optimierung ihrer Eigenschaften ist daher eine herausfordernde Aufgabe für die Wissenschaft. Gegenwärtig werden eine Vielzahl von Materialien zur Anwendung in Technologien zur sauberen Energie-

konversion aktiv weiterentwickelt. Heusler-Legierungen bilden eine dieser Materialklassen. Sie bestehen hauptsächlich aus nicht-seltenen Komponenten und besitzen multifunktionale Eigenschaften, die stark von der lokalen Anordnung der Elemente abhängen. Wir analysieren Gitterschwingungen, die magnetischen Eigenschaften und komplexen Ordnungsmechanismen in Ni-Mn-(In,Sn) Heusler-Legierungen mit Hilfe von Computer-Simulationen basierend auf ersten Prinzipien – das heißt ohne Rückgriff auf das Experiment. Unser Verfahren beruht auf der Dichtefunktionaltheorie, die eine akkurate und effiziente Charakterisierung der Beziehung zwischen Struktur, Magnetismus und elektronischen Eigenschaften ermöglicht. Unsere Ergebnisse demonstrieren, dass durch systematisch induzierte Unordnung, hervorgerufen durch Abweichungen von der Stöchiometrie oder Platzwechsellvorgänge zwischen den Elementen, ein Fein-Tuning der funktionellen Eigenschaften dieser Heusler-Systeme möglich ist. Dies eröffnet einen neuen Weg, Heusler-Materialien für die effiziente Anwendung im Bereich der Energiekonversion zu optimieren.

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Annotations

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