Full and half-Heusler compounds

Jason K. Kawasaki*, Shouvik Chatterjee, and Paul C. Canfield, Guest Editors

Heusler and half-Heusler compounds are ternary intermetallic compounds with highly tunable magnetic, topological, multiferroic, and electronic properties. They share a common crystal structure with more than 1000 members and compositions spanning most of the periodic table. This issue of *MRS Bulletin* covers the fundamental properties, synthesis science, and applications of Heusler and half-Heusler compounds from leading experts in theory and experiment. Topics include novel magnetic properties, thermoelectrics, skyrmions, spintronics applications, unconventional superconductivity, topological properties, and martensitic phase transitions. New approaches for materials discovery using high-throughput and big data approaches are highlighted.

What are Heusler compounds?

Heusler compounds are ternary intermetallics with more than 1000 members and compositions that span most of the periodic table. In these materials, the strong *d* character of bands near the Fermi energy as well as the variety of local and hybridized moments available in the 4*f*-shell give rise to highly tunable (and coupled) magnetic,[1–3] magnetoelastic,[4–6] topological,[7–11] superconducting,[9,12,13] heavy fermion,[14] and thermoelectric[15] properties. Many of these properties can be understood in terms of simple electron counting arguments.[16] The high degree of tunability in properties and compositions, and strong coupling to structural distortions, make Heusler compounds an exciting platform for materials design, both in bulk materials and at interfaces in epitaxial layered heterostructures.[17]

The cubic full and half-Heuslers are the most common structures in the Heusler family (**Figure 1**). Full Heusler compounds, with composition X_2YZ (or $XX'YZ$), crystallize in the cubic $L2_1$ structure, which consists of four interpenetrating face-centered-cubic (fcc) sublattices for the *X, X', Y*, and *Z* elements, respectively. Half-Heusler compounds have composition *XYZ* and are related by removal of the *X'* sublattice. Note that two common naming conventions exist for Heusler and related phases, which are related by switching the positions of *X* and *Y*. Here, we adopt the historical convention that is often used in metallurgy, where element *X* in the full Heusler structure is located at the unique lattice site with tetrahedral coordination to *Y* and tetrahedral coordination to Z. In this convention, full Heusler is denoted X_2YZ. This is the convention adopted by He et al.[18] in this issue. The other convention, commonly used in solid-state chemistry, is to order the elements in increasing electronegativity. In the electronegativity convention, full Heusler is XY_2Z, where *Y* is the dual tetrahedrally coordinated species. This is the convention adopted by Brod et al.[19] in this issue.

Other structural variations exist, which are characterized by structural distortions, variations in atomic site ordering, or by variations in atomic layer stacking. Starting from the full Heusler crystal structure, tetragonal distortions lead to the tetragonal Heuslers and are common in $X\mathrm{Mn}_2Z$ compounds. Such distortions are often Jahn–Teller-like and important because they can give rise to strong perpendicular magnetic anisotropy for applications in magnetic memory. Many full Heusler compounds also undergo martensitic phase transitions,[4] characterized by a shear or dilation of the cubic phase to a lower symmetry monoclinic or tetragonal phase. Starting from full Heusler, replacing the *X'* sublattice with *Y* sublattice leads to the inverse Heusler, which lacks the centrosymmetry of the original crystal lattice. Starting from the half-Heusler, which can be described by an *ABC–ABC* stacking of (111) planes (fcc), hexagonal Heusler compounds are characterized

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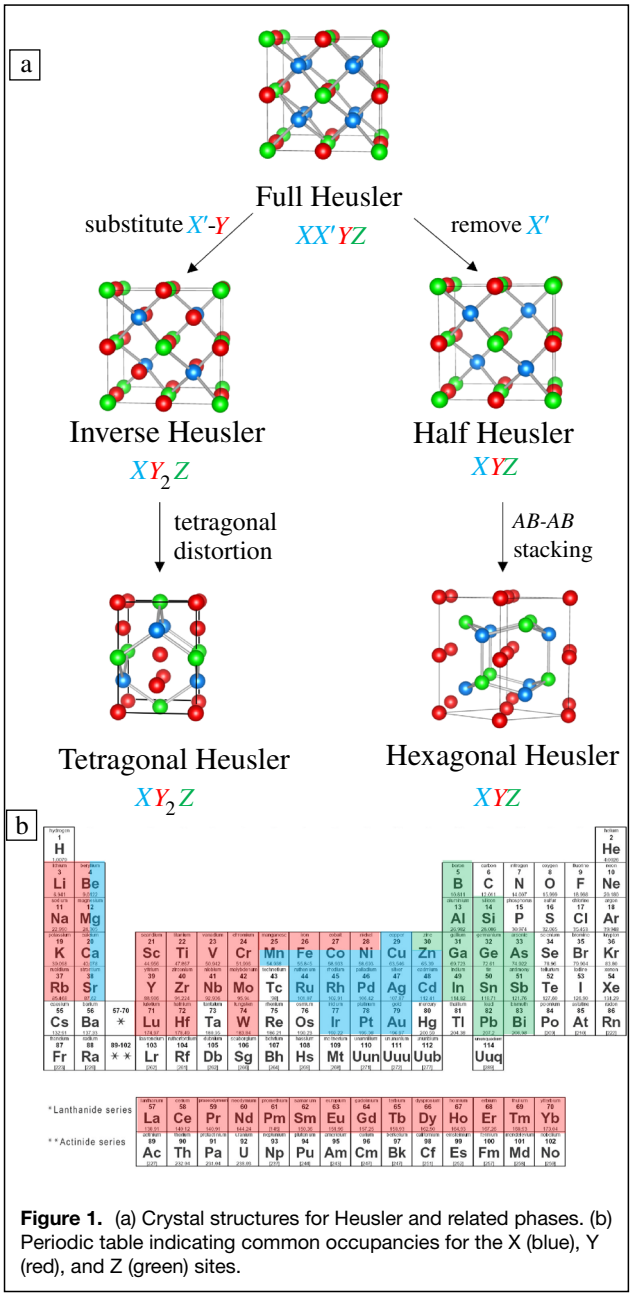

Figure 1. (a) Crystal structures for Heusler and related phases. (b) Periodic table indicating common occupancies for the X (blue), Y (red), and Z (green) sites.

by *AB–AB* stacking. Due to their unique polar axis, hexagonal Heuslers have been suggested as new ferroelectric materials.[20]

This issue: From magnetism to topology, phase transitions, thermoelectrics, and materials discovery

Significant interest in these compounds stems from magnetism and the strong coupling of magnetism to strain, structural transitions, and band topology. Heusler compounds were first discovered by Fritz Heusler in the 1900s. At the time, it was surprising to find ferromagnetism in an Heusler alloy despite it being composed of elements that are nonmagnetic in its elemental form. Since then, Heusler compounds have been at the forefront of research on magnetism.

One of the exciting properties that in principle could be realized in Heusler compounds is half-metallicity, which results in 100% spin polarization at the Fermi level. Along with high spin polarization, low Gilbert damping (damping torque for magnetic relaxation),[21] perpendicular magnetic anisotropy, high Curie temperature, and possible integration with technologically relevant III–V and Group IV semiconductors make Heusler compounds an attractive material choice for applications in spintronics. It is therefore not surprising that Heusler compounds have been extensively investigated for their potential as spin injectors and detectors in semiconductor devices operating at room temperature. In this issue, Hamaya et al.[22] summarizes recent progress in the development of Co-based full Heusler compounds for highly efficient spin transport both in GaAs and Ge-based devices. Here, the nature of the heterointerface between the Heusler compound and the semiconductor (GaAs/Ge) plays a crucial role in the device performance. Optimization of growth strategies and incorporation of judiciously chosen buffer layers could mitigate such issues that have resulted in the demonstration of two-terminal magnetoresistance of more than 0.1% at room temperature in semiconductor spintronic devices. Progress in the development of Heusler compounds and their efficient integration with semiconductor structures promise to make spin MOSFETS a viable technology in the near future.

Writing about another exciting research direction, Hirohata et al.[23] describes how the attractive magnetic properties of Heusler compounds could also be harnessed to realize spin-dependent transport resulting in large magnetoresistance ratios in GMR/TMR (giant/tunneling magnetoresistance) devices. They highlight the importance of composition controlled tunability of magnetic properties in Heuslers and the necessity of pristine interfaces, which have resulted in the record TMR ratio of 342% at room temperature (616% at 4 K) with $RA = 2.5 \times 10^3\ \Omega\ \mu m^2$ (product of resistance [R] and cross-sectional area of the device [A]) for epitaxial $B2$-Co_2FeAl/$Co_{0.5}Fe_{0.5}$/$MgAl_2O_4$ /$B2$-Co_2FeAl heterostructure. Because a number of elements including those with large atomic spin–orbit coupling (partially filled d and f shell) could be incorporated in the Heusler structure, Heusler compounds can efficiently generate large spin current via spin–orbit torque. Hirohata et al. summarizes these developments and provides a perspective on their applications in next-generation memory devices, spin caloritronics, magnetic sensors, and neuromorphic and stochastic computing.

Topology in condensed-matter systems is increasingly playing a central role in our understanding of material properties and in discovery of new phases. In carefully designed material systems, it is possible to realize topologically protected chiral magnetic textures known as skyrmions. Such topological magnetic defects could be manipulated by the application of electric field and are prime candidates for the realization of low-power racetrack memory.

In this issue, Felser and Parkin illustrate[24] how tunability in elemental composition, crystalline symmetry, spin–orbit coupling, and bulk noncentrosymmetry in certain Heusler compounds offers a unique material platform to realize skyrmion structures of different flavors such as Bloch, Néel, and antiskyrmions. Additional advantage in Heusler structures is the stability of skyrmions in a wide temperature-field range, including room temperature. The skyrmion size could also be controlled by modifying film thickness. Stabilization of more than one flavor of topological spin texture (viz., Bloch skyrmions and antiskyrmions) and their interconversion by the application of magnetic field/temperature has been recently achieved in $Mn_{1.4}PtSn$. The authors outline how further progress could be made in stabilization and manipulation of designer magnetic textures by exploiting unique properties of Heusler compounds.

Topology also has important consequences in momentum space. Mun and Bud'ko[25] describe the confluence of electronic band topology, heavy fermion behavior, and unconventional superconductivity in the half-Heusler *R*PtBi (*R* = rare earth) compounds. In these compounds, the strong band inversion, magnetism of the 4*f* electrons, and spin–orbit coupling lead to highly tunable properties. The authors review many of the key experimental signatures. For Weyl semimetals (e.g., GdPtBi), a negative longitudinal magnetoresistance is a key signature of the expected chiral anomaly. This family of compounds is typically antiferromagnetic, with either a [111] or [100] ordering vector as revealed by neutron diffraction. For heavy fermion behavior, hybridization of the 4*f* electrons varies significantly from CePtBi (small m*) to YbPtBi (large m* and quantum criticality). Finally, the modest superconducting transition temperatures ($T_c \sim 1$ K) despite the low carrier density, possible Cooper pairing with higher angular momentum, and line nodes in the superconducting gap have led to suggestions that YPtBi and LuPtBi may be topological superconductors.

The magnetism in many Heusler compounds also couples strongly to the structure. Heczko, Seiner, and Fähler[26] provide an introduction to ferromagnetic shape-memory alloys, in which ferromagnetism couples strongly with ferroelastic martensitic phase transitions. The full Heusler alloy Ni_2MnGa and its derivatives are representative of this class of materials. Across the magnetic-field-induced martensitic phase transition, these materials undergo strains as large as 12%, which is two orders of magnitude larger than giant magnetostriction. The authors review the broad family of related phenomena, including metamagnetic transitions, superelasticity, supermobility of twin boundaries, and applications for actuation, sensing, and energy conversion.

Half-Heusler compounds are attractive for thermoelectric energy conversion due to their large power factors, thermal and mechanical stability, and compositions that consist of inexpensive and Earth abundant elements. Brod et al. describe a strategy for enhancing the thermoelectric performance of semiconductor half-Heusler compounds, by enhancing the valley degeneracy in the electronic band structure.[19] The concept of avoided band crossings explains the orbital character at the band extrema (valleys). The valley degeneracy can be enhanced by tuning the electronegativity difference between the three elements, especially by alloying.

Finally, the wide range of compositions and tunable properties in Heusler compounds necessitates new approaches to systemically discover new materials with target properties. He et al. describe recent advances in computational materials discovery by high-throughput density functional theory calculations and machine learning.[18] The authors highlight recent successes in predicting compounds for thermoelectrics, half-metallic and spin-gapless ferromagnets, piezoelectrics, electrophotonics, and particle hardening. Common themes include integration with quantum materials databases and considerations of stability and metastability.

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Data availability

Data sharing not applicable to this article as no data sets were generated or analyzed during the current study.

Conflict of interest

The authors declare no conflict of interest.

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