

Compensated ferrimagnetic Heusler alloys: A search for the forgotten Néel's *L*-type ferrimagnetGerhard H. Fecher^{✉,*}, Shogo Yamashita[✉], Esita Pandey[✉], Atsufumi Hirohata^{✉,†} and Claudia Felser[✉]*Max-Planck-Institute for Chemical Physics of Solids, D-01187 Dresden, Germany*

(Received 28 November 2025; accepted 3 February 2026; published 3 March 2026)

In his groundbreaking work on the theory of ferri- and antiferromagnetism, Néel proposed the existence of ferrimagnets with zero net magnetization. These *L*-type ferrimagnets were later termed fully compensated. This study focuses on *L*-type ferrimagnets, particularly Heusler alloys based on vanadium (V), manganese (Mn), iron (Fe), and cobalt (Co). First-principles calculations reveal that these ferrimagnets exhibit anomalous temperature dependence, differing from antiferromagnets. Spin dynamics calculations, based on the calculated exchange coupling coefficients, show that this behavior stems from the ferrimagnets' inequivalent magnetic sublattices, which have distinct exchange interactions. The temperature dependence of the magnetic properties is analyzed through a comparison of the first-principles calculations and Néel's molecular field theory, extended to include Heusler compounds. In addition to exhibiting compensated ferrimagnetic behavior, examining the electronic structure using the Bloch spectral function shows that the investigated Heusler alloys are half-metals which have a full spin polarization at the Fermi energy. The combination of complete magnetic compensation and half-metallicity makes these alloys interesting for fundamental research and spintronics applications because their zero net magnetization minimizes stray fields while retaining half-metallicity.

DOI: [10.1103/913-4x5y](https://doi.org/10.1103/913-4x5y)**I. INTRODUCTION**

Néel, in his pioneering work on ferri- and antiferromagnetism, investigated several types of ferrimagnets that exhibit a compensation point [1]. The compensation point is the temperature at which the sublattice magnetizations are equal and the overall magnetization vanishes. In addition to the *N* and *V* types, he proposed an *L*-type ferrimagnet with a unique behavior. Specifically, the *N* and *V* types exhibit a compensation point, as well as zero or negative slopes of the magnetization $M(T)$ at $T = 0$, respectively. In contrast, an *L*-type ferrimagnet behaves similar to an antiferromagnet in that the magnetic moments of the sublattices compensate each other, resulting in zero net magnetization, at least at zero temperature (see Fig. 8 in Refs. [1,2] and Fig. 1 below). However, this *L*-type behavior was largely overlooked in later studies (see the Nobel Prize lecture [3] or Refs. [4,5]). The temperature dependencies of different types of compensated ferrimagnets, as discussed by Néel [1], are compared in Fig. 1. It is worth noting that the *L* type also appears for parameters other than those of the original Néel model, provided that they result in a vanishing total magnetization at zero temperature (see Appendix and Supplemental Material [6] for more information).

In general, ferrimagnets with two magnetic sublattices that are completely compensated exhibit an anomalous temperature dependence, which differs from that of antiferromagnets. Their magnetization vanishes at $T = 0$ but not between zero and the Néel (or Curie) point. This behavior arises from inequivalent magnetic sublattices with distinct exchange interactions. To maintain low total magnetization at the working point of a device, a high Curie temperature is required. To this end, it is desirable to identify multiautomic alloys in which the sublattice magnetizations are equal and independent of temperature. This requires magnetic materials with suitable exchange coupling parameters that enable such behavior.

Notably, although Néel's work [1] focused on ferrites, it mentioned the prototype Heusler compound Cu_2MnAl [7]. Heusler compounds exhibit a wide range of magnetic orderings and have been applied in spintronics owing to their simple crystal structure and electrical and magnetic properties that can be easily tuned by varying the stoichiometry. This is attributable to their simple crystal structure and electrical and magnetic properties that can be easily tuned by varying the stoichiometry (see, for example, Refs. [8] and [9]). Transition-metal-based compounds that crystallize in the $L2_1$ structure are typically ferromagnetic. However, those with *X* or *Y* structures are often ferrimagnetic. The different structures of Heusler compounds are illustrated in Fig. 2. In space group $F\bar{m}\bar{3}m$ (225), the occupied Wyckoff positions are $8c$ ($1/4, 1/4, 1/4$), $4b$ ($1/2, 1/2, 1/2$), and $4a$ ($0, 0, 0$). The $8c$ position is split into $4d$ ($3/4, 3/4, 3/4$) and $4c$ ($1/4, 1/4, 1/4$) in space group $F\bar{4}3m$ (216).

In special cases, the overall magnetic moment of a compound can vanish because the magnetic moments of the individual atoms cancel each other out in the ferrimagnetic state. This cancellation can occur either at zero temperature

*Contact author: gerhard.fecher@cpfs.mpg.de

†Also at Tohoku University, Center for Science and Innovation in Spintronics, Sendai 980-8577, Japan.

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI. Open access publication funded by Max Planck Society.

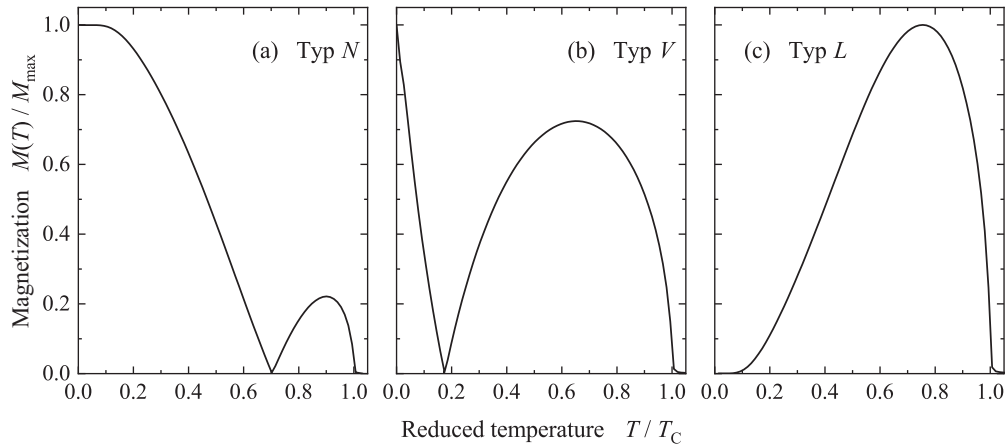


FIG. 1. Types of compensated ferrimagnets. (a) Type N : $dM/dT|_{T=0} = 0$. (b) Type V : $dM/dT|_{T=0} < 0$. (c) Type L : $dM/dT|_{T=0} = M(0) = 0$. The total magnetization is normalized to its maximum value, and the temperature is normalized to the Curie temperature T_C .

($T = 0$) or at a finite temperature referred to as the compensation temperature (T_{comp}). Compounds belonging to the first category ($T = 0$) correspond to Néel's L -type ferrimagnets and are termed completely compensated ferrimagnets. However, these compounds are sometimes incorrectly referred to as antiferromagnets, as demonstrated by symmetry arguments (see Supplemental Material for more information [6]).

In antiferromagnets, the density of states is identical for both spin channels, rendering them either insulating (semiconducting) or metallic. In contrast, ferrimagnets exhibit different densities of states for the two spin channels. This can result in a half-metallic state, in which one spin channel is conducting and the other is insulating [10,11]. Néel differentiated between (compensated) ferrimagnets and antiferromagnets by specifying that their magnetic sublattices are nonidentical and identical, respectively. Nevertheless, both systems exhibit negative interactions between the sublattices.

According to Ref. [12], the following criteria can be used to identify compensated and L -type ferrimagnetic Heusler compounds and alloys: (i) $n_v = 24 \pm \epsilon$ valence electrons and (ii) localized magnetic moment on the $4b$ (or $4a$) Wyckoff position.

Requirement (ii) is usually met when manganese (Mn) [or a rare-earth element, such as gadolinium (Gd)] occupies the

$4b$ site of space group 225 or 216, at least partially. The key role of the localized magnetic moment of Mn in Heusler compounds was initially investigated by Kübler *et al.* [13]. Completely compensated ferrimagnets occur when $\epsilon = 0$ (and thus, $n_v = 24$). In this case, the compensation point appears at $T = 0$. The compensation point can be tuned to finite temperatures when $\epsilon \neq 0$ but typically remains low depending on alloy composition (see Refs. [14,15]).

Completely compensated ferrimagnets have been observed in Heusler compounds with 24 valence electrons (n_v) [12,14,15]. The prototype proposed in Ref. [12] was Mn_3Ga . $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$ and $\text{Mn}_{2.5}\text{V}_{0.5}\text{Si}$ have also been identified as completely compensated half-metallic ferrimagnetic Heusler alloys. However, the Si-containing compound exhibited a low T_C . This effect was later demonstrated to occur in $\text{FeMn}_{1.5}\text{V}_{0.5}\text{Al}$, which has a Curie temperature exceeding 300 K.

To achieve ferrimagnetism in these 24-valence-electron Heusler compounds, a localized moment must be introduced [requirement (ii)]. Otherwise, the materials become narrow-gap semiconductors or semimetals that lack spontaneous magnetization. Examples of these materials include Fe_2VAl , CoFeTiAl , and CoMnVAl .

Many other Heusler alloys and compounds exhibit ferrimagnetic behavior involving two or three magnetic sublattices. To achieve high Curie temperatures, large magnetic moments are typically required [16,17]. Additionally, compounds with $n_v = 25$, such as CoMnCrAl (and variants with Ga or In instead of Al), exhibit high densities of states at the Fermi energy, which often results in unstable crystal structures. Mn_2CoAl and Mn_2CoGa , which have 26 valence electrons, are also ferrimagnetic. However, no compensation point has been observed in these spin-gapless semiconductors nor in similar compounds. The hypothetical compound CoMnCrSi is also ferrimagnetic and predicted to be close to a spin-gapless state.

A promising approach to identifying alternative completely compensated materials with 24 valence electrons and a localized magnetic moment is to start from well-known Mn-containing Heusler compounds. This design scheme, used in Ref. [12], resulted in the creation of $\text{MnFe}(\text{Mn}_{0.5}\text{V}_{0.5})\text{Al}$. This

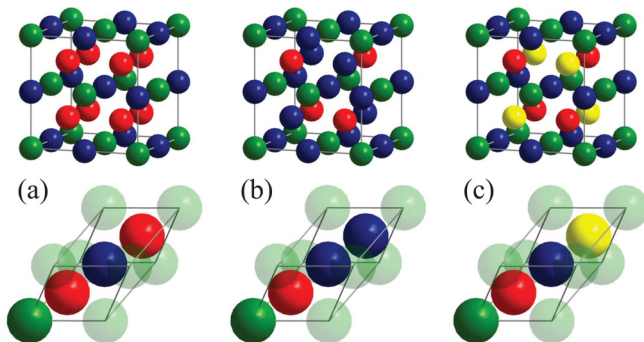


FIG. 2. Structure types of Heusler compounds. The figure shows the cubic and primitive cells for (a) $L2_1$: $Fm\bar{3}m$, (b) X : $F\bar{4}3m$, and (c) Y : $F\bar{4}3m$. Different atoms are distinguished by color.

alloy consists of Mn_2VAl ($n_v = 22$) and Fe_2MnAl ($n_v = 26$). Similarly, combining Mn_2VSi ($n_v = 21$) and Mn_3Si ($n_v = 25$) resulted in $\text{Mn}_2(\text{Mn}_{0.5}\text{V}_{0.5})\text{Si}$. In both cases, the completely compensated state originates from a 1 by 1 mixture. Ferrimagnets with a compensation point above $T = 0$ arise when the amount of the base compound with fewer valence electrons (here, 22) is slightly increased. Co-containing alloys, such as Mn_2VAl and Mn_2CoAl combinations, lead to the alloy $\text{Co}_{0.25}\text{Mn}_{0.75}\text{Al}$. While searching for such combinations, it is important to ensure that some Mn atoms occupy sites that lead to localized magnetic moments.

In addition to three-dimensional alloy systems, there is a growing interest in two-dimensional (2D) fully compensated ferrimagnets, which are a special type of 2D magnetic system (see, for example, Refs. [18,19]). Various fully compensated 2D ferrimagnets have recently been proposed by theoretical models [20,21]. Unlike the half-metals presented here, these systems are insulators [22]. It is worth noting that one of the first proposed three-dimensional fully compensated ferrimagnet systems (MnCrSb) [11,23] does not require alloying either, similar to the suggested 2D systems. However, it is difficult to synthesise it as a bulk material with the required $C1_b$ crystal structure. As with the proposed 2D systems, it may be possible to grow it in thin films or heterojunctions. In that case, one would have a quasi-2D half-metal that is fully compensated. Regarding alloy thin films, $\text{Mn}_2\text{Co}_{1-x}\text{V}_x\text{Al}$ has already been grown with a magnetic moment approaching zero at $x = 0.5$ [24]. This demonstrates the feasibility of using compensated ferrimagnetic half-metallic alloy systems in thin films for spintronic devices.

II. DETAILS OF THE CALCULATIONS

The electronic structure of the compounds was calculated within the nonlocal spin density approximation (NLSDA). Self-consistent electronic structure calculations for compounds with integer site occupations and alloys with non-integer site occupations were performed using the spin-polarized relativistic Korringa–Kohn–Rostoker method (MUNICH-SPRKKR) provided by Ebert *et al.* [25–27]. This program solves the Dirac equation in NLSDA based on either the atomic sphere approximation with spherical potentials or full-symmetry potentials. In this study, the atomic sphere approximation method was used only for initial tests, and all reported results were obtained using the full-potential mode. Notably, MUNICH-SPRKKR also incorporates the coherent potential approximation (CPA) [28] to calculate the properties of alloy systems with random distribution of the atoms.

All calculations were performed using one of the most common parametrizations of the exchange-correlation functional, that is, the generalized gradient approximation of Perdew *et al.* [29]. The CPA tolerance was set to 10^{-4} . The energy convergence criterion was set to 10^{-5} . f states were included in the basis set for all atoms. Integration was performed using 1240 ($F\bar{m}\bar{3}m$) or 2290 ($F\bar{4}3m$) irreducible k points, corresponding to a $36 \times 36 \times 36$ mesh. The density of states was calculated for the same number of k points using the Green's function with a small imaginary energy component of 2 mRy. Although the use of smaller values for the imaginary part can enhance the visibility of band gaps, they can also

TABLE I. Site occupations of alloys in the $D0_3$ or $L2_1$ structures (space group 225). Note that the degeneracy of the $8c$ position is twice that of the $4b$ position. Mn_3M compounds are presented for comparison. Consider the different amounts of Fe and Co. Note: Exchanging the occupations of the $4a$ and $4b$ sites does not change the structures.

	8c	4b	4a
Mn_3M	Mn	Mn	$M = \text{Al, Ga}$
$\text{Mn}_{2.5}\text{V}_{0.5}M$	Mn	Mn + V	$M = \text{Si, Ge, Sn}$
$\text{Mn}_{1.5}\text{FeV}_{0.5}M$	Mn + Fe	Mn + V	$M = \text{Al, Ga}$
$\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}M$	Mn + Co	Mn + V	$M = \text{Al, Ga}$

increase the noise in the density of states. The imaginary part introduces a Lorentzian-type broadening of the states that leads to a finite density of states across the minority band gap. Therefore, the presence of a minority band gap was verified by supercell calculations (see Ref. [12]). Before calculating the properties, the lattice parameters were optimized for each material and composition in the full-potential mode (see the Supplemental Material [6]).

To visualize the electronic structure, Bloch spectral functions [30] were calculated in the fully relativistic mode. This mode of MUNICH-SPRKKR solves the spin-polarized Dirac equation and, when combined with CPA, allows one to explore the electronic structures in systems without periodicity in the atomic arrangement within the mean-field framework. In most previous studies, half-metallicity was demonstrated only for well-ordered compensated ferrimagnetic compounds (e.g., Ref. [12]), whereas the corresponding alloys were modeled using artificial supercells, which do not reflect the random distribution of the atoms. In practice, random site occupations in alloys destroy the periodicity of the atomic arrangement, even if the lattice itself remains periodic. Consequently, the usual band structure is no longer well-defined. Therefore, the Bloch spectral function $[E(k)]$, which can be interpreted as a momentum (k)-resolved density of states, was calculated to verify the half-metallicity of the investigated materials.

Magnetic phase transition temperatures (T_C^{MF}) were calculated from the Heisenberg exchange integrals, following the method of Liechtenstein *et al.* [31] for local spin density approximation calculations. The extension of this approach to multisublattice systems has been described by Anderson [32]. Details of the determination of the exchange coupling constants J_{ij} using MUNICH-SPRKKR can be found in Refs. [33–35]. The cluster size was set to 4 to 5 times the lattice parameter, yielding up to 1156 ij combinations of the exchange coupling parameters $J_{ij}(r)$ for each atom.

The CPA method was used to model the statistical distribution of transition metals on a common site of the $L2_1$ crystalline structure for alloys with noninteger site occupations. The transition metals were assigned to the $8c$ and $4b$ sites, while the main-group element occupied the $4a$ position. Representative site occupations are listed in Table I.

The calculated site-resolved magnetic moments and averaged exchange coupling coefficients were subsequently used in VAMPIRE 6.0 to determine the temperature dependence of the magnetization using Monte Carlo methods and atomistic

TABLE II. Site occupations of compounds with X and Y structures (space group 216). Note the different amounts of Fe and Co. Note: Exchanging pairwise the occupations of sites $4a$ and $4b$, $4d$ and $4c$, or both groups does not change the structures.

		$4d$	$4c$	$4b$	$4a$
Mn_2CoM	X	Co	Mn	Mn	$M = \text{Al, Ga}$
CoMnCrM	Y	Co	Mn	Cr	$M = \text{Si, Ge}$
$\text{Mn}_{1.5}\text{FeV}_{0.5}M$	Y	Mn	Fe	Mn + V	$M = \text{Al, Ga}$
$\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}M$	Y	Mn	Mn + Co	Mn + V	$M = \text{Al, Ga}$

spin dynamics [36]. A mesh with dimensions of approximately $20 \times 20 \times 20$ was used, corresponding to more than 1.1×10^5 magnetic atoms. Any possible magnetic polarization of the main-group elements was neglected in these calculations, that is, they were not included in the calculations. Typically, the Curie temperatures are overestimated by the mean field values T_C^{MF} calculated directly from the exchange coupling coefficients. Therefore, the magnetic transition temperatures (T_C^{SD}) were additionally taken from a fit of the magnetization of the dominant Mn atom on $4b$ to a $(1 - T/T_C)^\beta$ law [36,37] because a fit of the total magnetization is not possible for the compensated ferrimagnets. The average uncertainty of these transition temperatures is $\Delta T_C = \pm 3.5$ K. The transition temperatures were cross-checked by comparison to the maximum of the susceptibility close to T_C , a phenomenon known as the Hopkinson effect [38,39].

Compounds with integer site occupations were additionally investigated using WIEN2K [40] to verify crystal parameters, minority-spin band gaps, and total magnetic moments. The integer site occupations of representative compounds with X_a or Y structures are listed in Table II. The alloys $\text{Mn}_{1.5}\text{FeV}_{0.5}M$ and $\text{Mn}_2\text{CoV}_{0.50.5}M$ may appear in either the $L2_1$ - or Y -type structure and are thus included in both Tables I and II. The imbalance in the occupation of the $4d$ and $4c$ sites may vary until a balanced stoichiometry of the $8c$ sites in $L2_1$ is reached. In case of equal occupations at the $8c$ and $4b$ sites, a disordered DO_3 -type structure forms. However, such structure types, as well as disordered $B2$ or mixed $L2_1 + Y$ structures, are beyond the scope of the present work.

Thus far, the only transition metals identified as constituents of the investigated Heusler alloys are V, Mn, Fe, and Co. Further combinations, also including other 3d elements, that could result in completely compensated half-metals are suggested in the supplement [6].

Notably, the presence of magnetization alters the crystal symmetry [41]. Starting from the $L2_1$ structure with space group $Fm\bar{3}m$ (225), the magnetic space group becomes $I4/m\bar{m}'m'$ (139.537) or $R\bar{3}m'$ (166.101) when the magnetization is along $[001]$ or $[111]$, respectively. For structures with $F\bar{4}3m$ (216), the corresponding magnetic space groups are $I\bar{4}m'2'$ (119.319) or $R\bar{3}m'$ (160.67) when the magnetization is along $[001]$ or $[111]$, respectively. Here, $'$ represents the spin-reversal operator; consequently, m' is a mirror operation followed by spin reversal. The same convention applies to the twofold rotation that becomes $2'$ when followed by spin reversal (see Refs. [42,43] for the appropriate notation and numbering of magnetic space groups).

TABLE III. Calculated magnetic properties of well-ordered ferrimagnetic Heusler compounds with nonvanishing total spin moments. Mn_3Al with a hypothetical DO_3 structure and vanishing moment is included for comparison (Note: In case of Mn_3Al , the $8c$ position of space group 225 was split into $4d$ and $4c$ of 216). The Curie temperatures (T_C^{MF}) are determined using the mean-field approach, directly from the exchange coupling coefficients. T_C^{SD} are the values from spin dynamic calculations.

	$m [\mu_B]$				T_C^{MF} [K]	T_C^{SD} [K]
	$4d$	$4c$	$4b$	Tot		
Mn_3Al	-1.5	-1.5	3.0	0.0	726	670
CoMnCrAl	0.7	-1.2	1.5	1.0	772	506
CoMn_2Al	1.0	-1.8	2.8	2.0	1122	986
CoMnCrSi	1.0	-0.8	1.8	2.0	734	506

III. RESULTS AND DISCUSSION

The calculated magnetic properties for various ferrimagnetic Heusler compounds and alloys are summarized in Tables III and IV, respectively. In addition to the hypothetical Mn_3Al , Table III lists the properties of conventional ferrimagnets. The moment of Mn on site $4c$ is aligned antiparallel to the moments on the other sites, thereby reducing the total moment. The difference with the hypothetical compensated ferrimagnet Mn_3Al is obvious. The largest moment always appears at the atoms occupying the $4b$ position. However, these Co-containing ferrimagnets (with three different magnetic sublattices) do not exhibit a compensation point.

Table IV lists the properties of completely compensated Néel L -type ferrimagnets with the $L2_1$ structure. In all cases, the total magnetic moment vanishes because the directions of the averaged magnetic moments of atoms on $8c$ and $4b$ sites

TABLE IV. Calculated magnetic properties of compensated Heusler alloys with vanishing total spin moments. The alloy properties were calculated for the $L2_1$ structure. Mn_3Al and Mn_3Ga with the DO_3 structure are included for comparison. Site-resolved magnetic moments are given per atom, and site occupation factors are not included. In all cases, the total spin magnetic moment is zero or approaches zero. Note the different amounts of Fe and Co in the respective alloys.

	$m [\mu_B]$				T_C^{MF} [K]	T_C^{SD} [K]
	$8c$	$4b$				
Mn_3M	Mn	Mn				
$M = \text{Al}$	-1.5	3.0			726	670
$M = \text{Ga}$	-1.6	3.2			702	617
$\text{Mn}_{2.5}\text{V}_{0.5}M$	Mn	Mn	V			
$M = \text{Si}$	-0.8	2.7	0.5		362	389
$M = \text{Ge}$	-0.9	3.0	0.6		419	259
$\text{Mn}_{1.5}\text{FeV}_{0.5}M$	Mn	Fe	Mn	V		
$M = \text{Al}$	-1.4	-0.3	2.9	0.5	411	395
$M = \text{Ga}$	-1.5	-0.3	2.9	0.6	424	368
$\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}M$	Mn	Co	Mn	V		
$M = \text{Al}$	-1.4	0.5	2.9	0.7	638	448
$M = \text{Ga}$	-1.5	0.6	3.1	0.8	666	494

are opposite. Since the $8c$ sites have twice as many atoms as the $4b$ sites, the occupation of these sites by Mn, Fe, or Co must be balanced appropriately. Splitting the $8c$ position yields three magnetic sublattices, two of which are equal. As in regular ferrimagnets, the largest magnetic moment always occurs at the $4b$ position, not only at Mn but also across different compositions.

A. Compounds with $n_v = 24$ and a ground state without magnetic order

Various ordered compounds with 24 valence electrons were tested using WIEN2K. For all the tested compounds, the spin-polarized calculations converged to a non-spin-polarized ground state, that is, both the total and individual magnetic moments vanished. Examples of compounds with a $L2_1$ structure and $n_v = 24$ are Fe_2TiSi [44], Fe_2VAl [45], Fe_2VGa [46], and Co_2ScAl . In these compounds, no localized magnetic moments are carried by the Sc, Ti, or V atoms, and neither Fe nor Co contribute magnetic moments to support a magnetic order. Fe_2VAl , which is stable in the cubic structure, resembles a narrow-gap semiconductor or semimetal. Although the existence of Mn_2CrSi has been predicted, its experimental proof remains to be reported. Quaternary Heusler compounds such as CoFeTiAl and CoMnVAl adopt a Y -type structure [47]. Despite the strongly ferromagnetic CoFe complex, CoFeTiAl exhibits no ferrimagnetic order or magnetic moments and thus also resembles a narrow-gap semiconductor or semimetal.

Hypothetical compounds with the X -type structure include Cr_2CoAl , Cr_2FeSi , and other similar systems with other main-group elements. Most of these compounds have not yet been synthesized. They are likely unstable and decompose into elements and binaries. Cr_2CoGa , proposed in Ref. [48], may exist but exhibits a high degree of disorder. Its X -type structure has not been proven experimentally [49]. The stability of other hypothetical compounds with a Y -type structure, such as FeMnCrAl and FeMnCrGa , remains unvalidated as well. In general, alloys predicted only from calculations must be regarded with caution, as most of them have never been synthesized.

B. Ferrimagnetic compounds with $n_v \neq 24$

In Heusler compounds and alloys with $n_v \neq 24$, a magnetic ground state with a magnetic moment of $m \approx |n_v - 24|$ is expected. This behavior has been observed across half-metallic ferromagnets. Well-known compounds include Mn_2CoM ($M = \text{Al}, \text{Ga}$) and the hypothetical compounds CoMnCrM ($M = \text{Al}, \text{Si}$), which are ferrimagnetic (see Table III). However, none of them exhibit a compensation point. The temperature dependence of the magnetization for Mn_2CoAl and the hypothetical compound CoMnCrAl (X - and Y -type structures, respectively) is presented in the Supplemental Material [6].

C. Completely compensated ferrimagnetic compounds and alloys: The Néel L type

The compounds Mn_3M ($M = \text{Al}, \text{Ga}$) become completely compensated when they adopt the $D0_3$ structure. At $T = 0$, the magnetic moment is zero because the magnetic moments

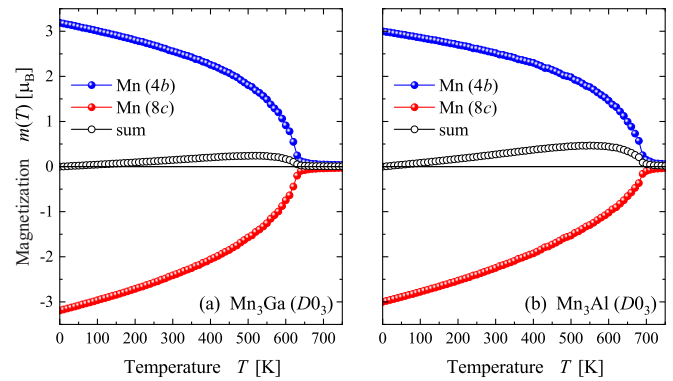


FIG. 3. Temperature-dependent magnetization of Mn_3Ga (a) and hypothetical Mn_3Al (b).

at the $8c$ and $4b$ sites cancel each other out. Details of the electronic structure of Mn_3Ga can be found in Ref. [12]. The calculated temperature dependence of the magnetization for Mn_3Ga is shown in Fig. 3(a), while its comparison with the hypothetical Mn_3Al is presented in Fig. 3(b). The site-resolved magnetization curves exhibit different temperature dependencies. Therefore, the total magnetization does not remain zero with increasing temperature. Instead, it reaches a maximum of approximately $0.2 \mu_B$ for Mn_3Ga and $0.6 \mu_B$ for Mn_3Al . The Curie point from the Monte Carlo spin dynamics calculations (Fig. 3) is slightly higher for Mn_3Al than for Mn_3Ga , which is consistent with the mean-field Curie temperature calculated directly from the exchange coupling constants (Tables III and IV).

The $\text{Mn}_{2.5}\text{V}_{0.5}M$ ($M = \text{Si}, \text{Ge}$) compounds are also completely compensated (Table IV). The calculated temperature dependence of the magnetization is illustrated in Fig. 4. The behavior is similar to that of Mn_3Ga , albeit with smaller values for the sublattices. This decrease is attributable to the lower average magnetization at the $4b$ site (occupied by Mn and V), as well as that of Mn at $8c$. The lower magnetic moment at Mn on $8c$ is a direct consequence of the lower averaged moment of the atoms on $4b$. As expected for a Néel L -type completely compensated ferrimagnet, the total magnetization vanishes at 0 K and exhibits a maximum of approximately $0.3 \mu_B$ near room temperature (300 K).

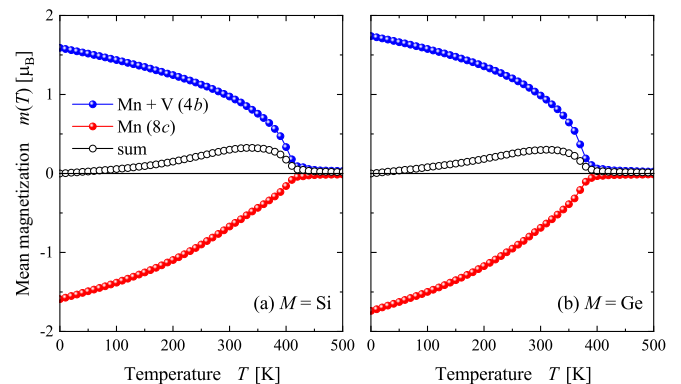


FIG. 4. Temperature-dependent magnetization of $\text{Mn}_{2.5}\text{V}_{0.5}\text{Si}$ (a) and $\text{Mn}_{2.5}\text{V}_{0.5}\text{Ge}$ (b).

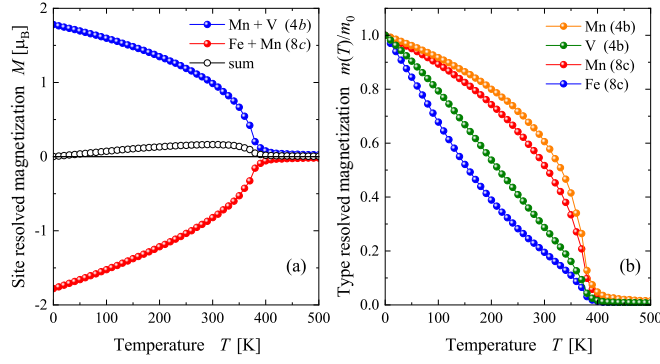


FIG. 5. Temperature-dependent magnetization of $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Ga}$. (a) shows the site (8c, 4b) resolved magnetization and (b) shows the type (Fe, Mn, V) resolved magnetic moments normalized to the 0 K values.

Both computations and experiments reveal that $\text{FeMn}_{1.5}\text{V}_{0.5}\text{Al}$ is a completely compensated or Néel L -type ferrimagnet. The same conclusion is derived through calculations in which Al is replaced by Ga. Figure 5 shows the calculated temperature dependence of the magnetization for $\text{FeMn}_{1.5}\text{V}_{0.5}\text{Ga}$. The maximum magnetization of $0.15\mu_B$ and T_C appear at slightly higher temperatures compared with the $\text{Mn}_{2.5}\text{V}_{0.5}\text{M}$ compounds. The sublattice magnetizations are nearly identical, confirming that the magnetic behavior is dominated by the localized moments on the $4b$ position. The atom-resolved magnetic moments are shown in Fig. 5(b). For better comparison, the magnetic moments are normalized to their values at 0 K. The most dominant moment is associated with Mn at the $4b$ position.

In $\text{FeMn}_{1.5}\text{V}_{0.5}\text{M}$ ($M = \text{Al}, \text{Ga}$) compounds, the Fe and Mn atoms do not necessarily occupy the L_{21} 8c position. Instead, they can occupy the $4d$ and $4c$ positions of the Y -type structure separately. The results for the two possible structures are presented in Table V and Fig. 6 for $\text{FeMn}_{1.5}\text{V}_{0.5}\text{Al}$. In the L_{21} structure, anti-site disorder occurs between Fe and Mn on the $4d$ and $4c$ sites of the Y structure (see Tables I and II). In both cases, Mn and V are randomly distributed on the $4b$ site. The mean-field Curie temperature derived from the exchange coupling coefficients is considerably higher in the Y structure with less disorder than in L_{21} , whereas the magnetic moments remain nearly unchanged.

TABLE V. Calculated magnetic properties of $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$ for different face-centered cubic structure. In Y , disorder appears only through the random occupation of $4b$ by Mn and V. In L_{21} , anti-site disorder occurs between Fe and Mn on 8c.

Structure	$m [\mu_B]$		T_C^{MF} [K]	T_C^{SD} [K]
L_{21}	8c	4b	411	396
	Mn + Fe	Mn + V		
	-1.4, -0.3	2.9, 0.5		
Y	4d	4c	519	388
	Mn	Fe		
	-1.4	-0.4		
		Mn + V		
		2.9, 0.7		

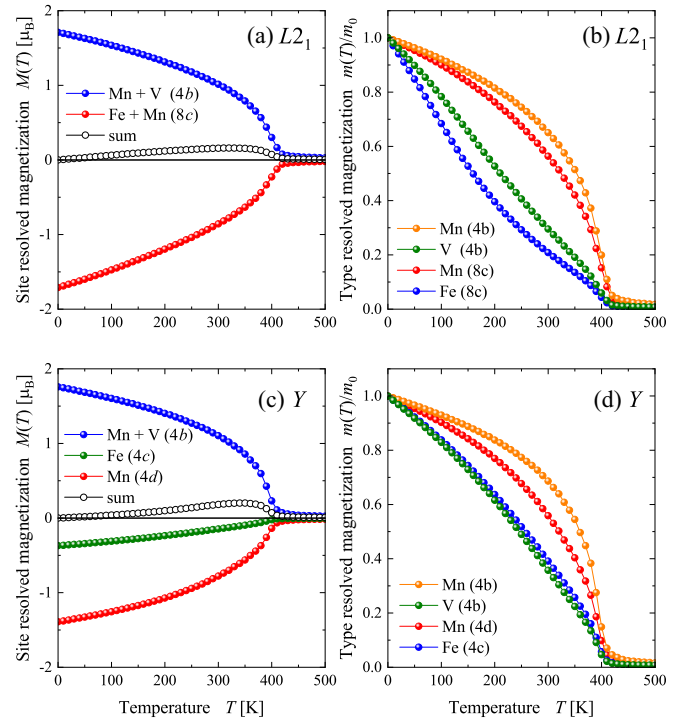


FIG. 6. Temperature-dependent magnetization of $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$. (a), (b) L_{21} - and (c), (d) Y -type structures. (a), (c) Site-resolved and (b), (d) type (Fe, Mn, V)-resolved magnetic moments. The latter are normalized to their 0 K values.

An alternative alloy with 24 valence electrons arises from a 1:1 combination of Mn_2VAl and Mn_2CoAl , both of which are stable Heusler compounds. The resulting alloy is $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$. The number of valence electrons remains the same when Al is replaced by Ga. Alloys with compositions $\text{Mn}_{2-\delta}\text{Co}_{0.5-\delta}\text{V}_{0.5}\text{Al}$ and $\text{Mn}_{2-z}\text{Co}_{0.5}\text{V}_{0.5-z}\text{Al}$ were investigated by Gavrea *et al.* [50], and those with compositions $\text{Mn}_{2-2x}\text{Co}_{0.5+x}\text{V}_{0.5+x}\text{Ga}$ were explored by Seredina *et al.* [51]. Notably, these studies did not report the presence of a completely compensated state, suggesting a considerable deviation from $n_v = 24$.

The calculated magnetic properties of $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$ for L_{21} and Y structures are summarized in Table VI. The site-resolved magnetic moments are comparable to those of the Mn-Fe-V-Al or-Ga systems. The advantage of $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$ is its higher Curie temperature relative to the

TABLE VI. Calculated magnetic properties of $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$ for different face-centered cubic structure. In Y , disorder arises from the random occupation of $4c$ by Mn and Co and $4b$ by Mn and V. In L_{21} , additional antisite disorder occurs between Co and Mn on 8c.

Structure	$m [\mu_B]$		T_C^{MF} [K]	T_C^{SD} [K]
L_{21}	8c	4b	638	449
	Mn + Co	Mn + V		
	-1.4, +0.5	2.9, 0.7		
Y	4d	4c	662	540
	Mn	Mn + Co		
	-1.4	-1.4, +0.6		
		Mn + V		
		2.9, 0.7		

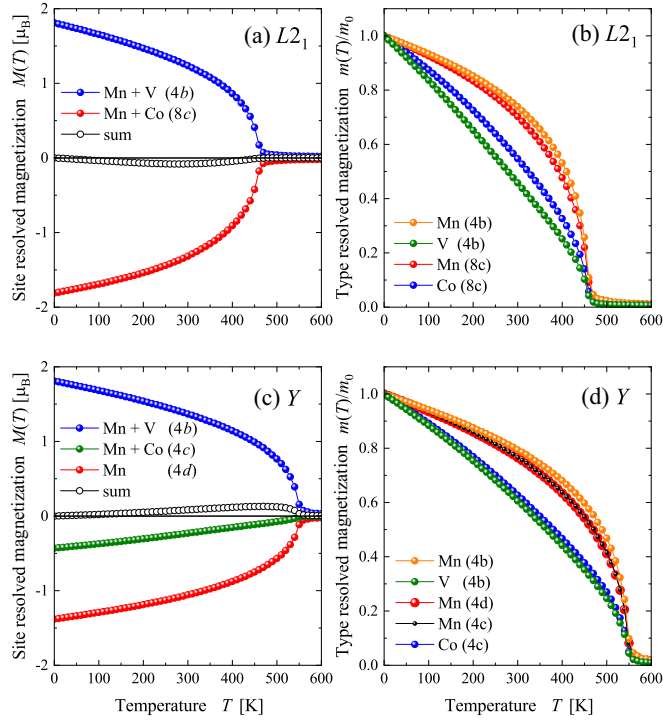


FIG. 7. Temperature-dependent magnetization of $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$. (a), (b) $L2_1$ - and (c), (d) Y -type structures.

Fe-containing alloy. Depending on the structure, T_C is calculated to be approximately 650 K, which is close to the value derived from experiments on off-stoichiometric alloys [50].

The key difference with the Fe-containing alloy is that the Co magnetic moments are opposite those of Mn at 8c or (4d, 4c). Figure 7 compares the thermomagnetic curves for $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$ with $L2_1$ - and Y -type structures. For the $L2_1$ -type structure, in particular, the temperature dependence trends of the local magnetic moments at Co and Fe are clearly different.

In all thermomagnetic plots of the L -type ferrimagnets, a maximum of the magnetization (m_{max}) occurs at a specific temperature (T_m). The magnitude and position of these maxima are summarized in Table VII. In most cases, T_m is close to room temperature (300 K). The lowest maximum magnetization is obtained for $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$.

The most interesting behavior occurs in the $L2_1$ structure of $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$. The total magnetic moment is opposite

TABLE VII. Maxima of the magnetization for various L -type ferrimagnetic Heusler alloys.

Compound	Structure	T_m [K]	m_{max} [μ_B]
$\text{Mn}_{2.5}\text{V}_{0.5}\text{Si}$	$L2_1$	340	0.324
$\text{Mn}_{2.5}\text{V}_{0.5}\text{Ge}$	$L2_1$	310	0.300
$\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$	$L2_1$	320	0.159
$\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$	Y	350	0.202
$\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Ga}$	$L2_1$	300	0.162
$\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$	$L2_1$	280	-0.081
$\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$	Y	470	0.129

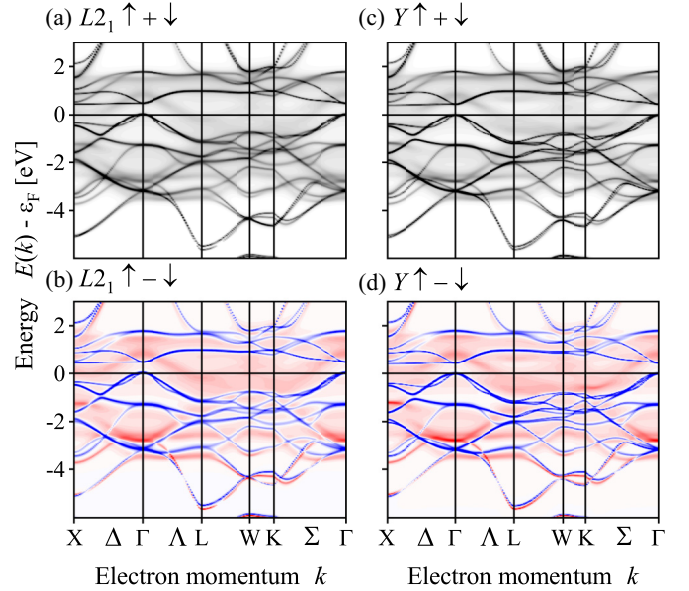


FIG. 8. Electronic structure of $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$ with $L2_1$ (a), (b) and Y (c), (d) -type structures. (a), (c) Sum of the Bloch spectral functions and (b), (d) the difference between the two spin channels. The latter plots correspond to the unnormalized spin polarization. The assignment of $\uparrow$ and $\downarrow$ for majority and minority states is derived for $T > 0$, using the magnetic moment of Mn on the 4b site.

the dominant magnetic moment at Mn on the 4b site. This is in contrast to the magnetic properties in the Y structure. These results suggest that near-complete compensation could be achieved within the temperature range up to T_C by altering the alloy composition.

Figure 8 shows the spin-resolved Bloch spectral function of $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$ with $L2_1$ - and Y -type structures. The magnetic moment vanishes at $T = 0$, and the number of electrons in the two spin channels is equal. This makes it difficult to assign majority and minority states. Therefore, the spin states $\uparrow$ and $\downarrow$ are derived from the majority and minority states of Mn on the 4b site. These states also govern the temperature dependence of the magnetization for $0 < T < T_C$. No significant differences are found between the centrosymmetric $L2_1$ and non-centrosymmetric Y structures. Only one crossing point at W ($E_W \approx -4.3$ eV) is clearly resolved. This point exists in $L2_1$ but is lifted in Y for both spin channels. Chemical disorder scattering affects only one spin channel owing to the random distribution of atoms on the 8c and 4b sites in the $L2_1$ alloy or their distribution only on 4b in the Y -type structure. The other channel exhibits a clear direct bandgap of approximately 750 meV at the Γ point for both structures. These results confirm that the half-metallic state is preserved in the alloys with random site occupations, as previously reported [14].

The situation is less clear in the case of $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Ga}$. Regardless of the cubic structure type, some minority bands shift above the Fermi energy. This destroys the half-metallicity. Such behavior is often observed when comparing Heusler compounds with Ga and Al. The larger lattice parameter in Ga compounds decreases the size of the Brillouin zone, resulting in a stronger dispersion of the bands.

The electronic structure of $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$ is illustrated in Fig. 9 using the spin-resolved Bloch spectral functions for

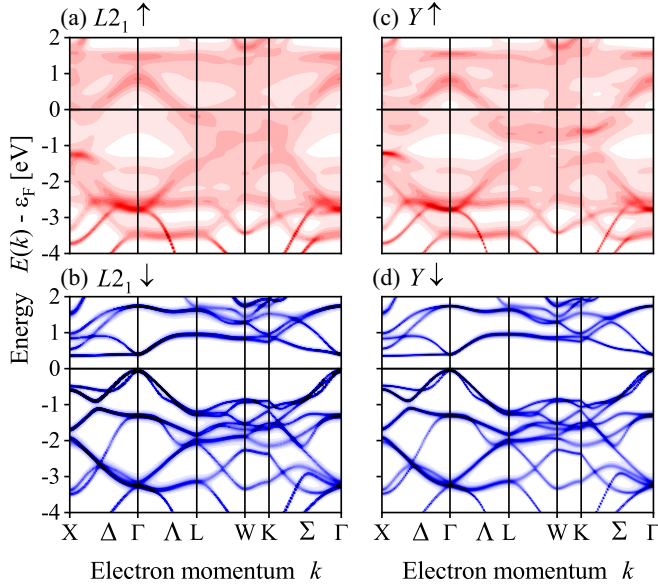


FIG. 9. Electronic structure of $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$ with (a), (b) $L2_1$ and (c), (d) Y -type structures. The assignment of $\uparrow$ and $\downarrow$ for majority and minority states is taken for $T > 0$ and the magnetic moment of Mn on the $4b$ site.

the $L2_1$ - and Y -type structures. Similar to $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$, the random distribution of atoms on the lattice sites primarily affects the majority states. The electronic structure is nearly independent of the actual crystal structure. The minority states exhibit a direct band gap of approximately 400 meV at the Γ point, confirming the half-metallic character.

D. Potential route to room-temperature compensated ferrimagnetic, half-metallic alloys

For practical applications, materials that remain compensated at or above room temperature are required. Two quantities must be optimized: Curie temperature and compensation temperature.

Figure 10 shows two examples of how the total magnetic moment at 0 K affects the magnetization curves $m(T)$. The magnetic moment is tuned by varying the Mn and V

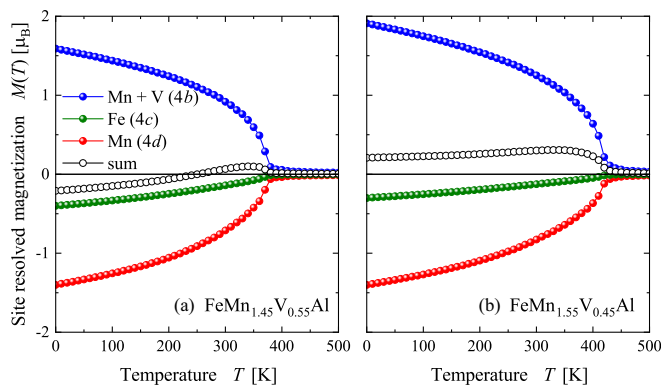


FIG. 10. Temperature-dependent magnetization of $\text{FeMn}_{1.5+x}\text{V}_{0.5-x}\text{Al}$. (a) undercompensated $x = -0.05$ and (b) overcompensated $x = +0.05$ at $T = 0$.

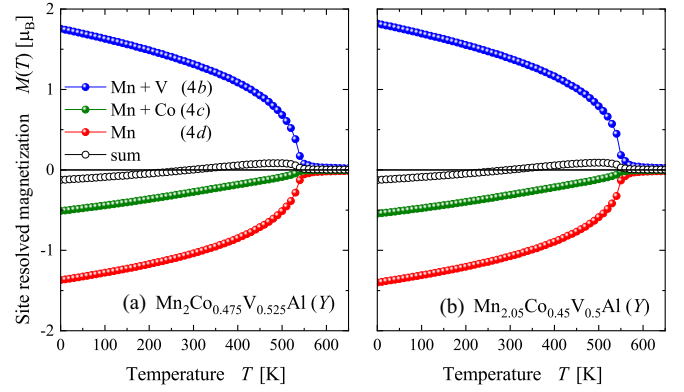


FIG. 11. Temperature-dependent magnetization in Mn-Co-V-Al alloys with $n_v \leq 24$ and Y -type structure. (a) $\text{Mn}_{2.1}\text{Co}_{0.45}\text{V}_{0.45}\text{Al}$ and (b) $\text{Mn}_{2.05}\text{Co}_{0.45}\text{V}_{0.5}\text{Al}$.

contents in $\text{Mn}_{1.5+x}\text{FeV}_{0.5-x}\text{Al}$. The magnetization changes with x by $2x \mu_B$. For $x < 0$, the sublattice moments are undercompensated ($n_v < 24$, $m < 0$) at 0 K, whereas they are overcompensated ($n_v > 24$, $m > 0$) at $x > 0$.

The temperature dependence of the total and sublattice magnetizations in Fig. 10 corresponds to that shown in Fig. 4 of Néel's work [1]. However, the zero crossings of the total magnetization are not displayed in Fig. 7 of Ref. [1]. The curves in the latter figure show the absolute value of the magnetization, characteristic of measurements under an applied magnetic field. The under- and overcompensated alloys represent Néel's N - and P -type ferrimagnets, respectively [1]. This is in agreement with measurements, where depending on the composition, transitions from P to L to N -type ferrimagnets were observed [14,15].

P - and N -type ferrimagnetic order was also reported by Gavrea *et al.* [50] for two different compositions in the Mn-Co-V-Al system. These measurements, performed under a high magnetic field (0.5 T), did not reveal a zero crossing at the compensation point, unlike the observations of Stinshoff *et al.* [14,15] for different compositions in the Mn-Fe-V-Al system. Two examples of Mn-Co-V-Al ferrimagnetic alloys exhibiting a compensation point are shown in Fig. 11.

So far, compensation points have been consistently observed when the number of valence electrons is slightly below 24. Prior experimental studies [14,15] showed that the positions of these points depend on the stoichiometry. Table VIII summarizes the calculated Curie and compensation points of selected alloys with less than 24 valence electrons. The compensation temperature approaches room temperature with increasing mean-field Curie temperature. The compensation temperature depends on the structure and composition.

TABLE VIII. Curie and compensation points (T_{comp}) of ferrimagnetic Heusler alloys with $n_v = 23.9$.

Compound	Structure	T_C^{MF} [K]	T_C^{SD} [K]	T_{comp} [K]
$\text{Mn}_{1.45}\text{FeV}_{0.55}\text{Al}$	$L2_1$	404	377	250
$\text{Mn}_{1.45}\text{FeV}_{0.55}\text{Al}$	Y	498	365	250
$\text{Mn}_2\text{Co}_{0.475}\text{V}_{0.525}\text{Al}$	Y	663	533	290
$\text{Mn}_{2.05}\text{Co}_{0.45}\text{V}_{0.5}\text{Al}$	Y	671	545	300

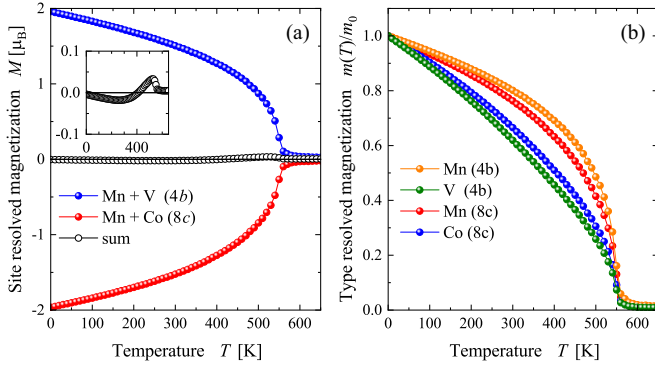


FIG. 12. Temperature-dependent magnetization of $\text{Mn}_{2.1}\text{Co}_{0.45}\text{V}_{0.45}\text{Al}$ with $L2_1$ structure. The inset in (a) shows the total magnetization on an enlarged scale.

E. L -type half-metallic ferrimagnet with additional compensation point

The Heusler alloys with $L2_1$, X , or Y structures are not strictly two-sublattice ferrimagnets. In particular, those with space group $F\bar{4}3m$ (216) may have three different magnetic sublattices owing to the four inequivalent Wyckoff positions. In systems with more than two magnetic sublattices, multiple compensation points can occur. Prussian blue and its analogs are typical examples of such ferrimagnetic ordering [52]. Néel has already stated that the generalization of the two sublattice theory of ferrimagnetism to three (or more) sublattices poses no problem [3].

In this study, a special behavior is observed for $\text{Mn}_{2.1}\text{Co}_{0.45}\text{V}_{0.45}\text{Al}$. This alloy has 24 valence electrons and is compensated at $T = 0$, as is typical for L -type ferrimagnets. However, a second compensation point appears at higher temperatures ($T_{\text{comp}} \approx 400$ K). The temperature dependence of the magnetization is shown in Fig. 12. The maximum magnetization is only $\approx 0.03 \mu_B$ at 510 K. A similar anomalous behavior of magnetization has been observed in low fields for $\text{Sm}_{1-x}\text{Gd}_x\text{Al}_2$, which was described by the authors as a zero-magnetization ferromagnet [53].

The Bloch spectral function of the $\text{Mn}_{2.1}\text{Co}_{0.45}\text{V}_{0.45}\text{Al}$ alloy with the $L2_1$ structure is shown in Fig. 13. The $E(k)$ relation is similar to that of $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Al}$ and is only weakly influenced by the slightly different stoichiometry. The clear

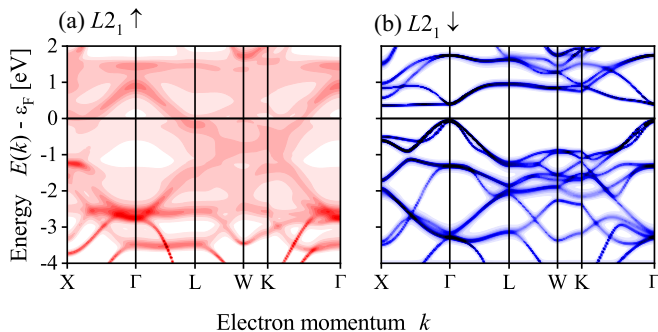


FIG. 13. Electronic structure of $\text{Mn}_{2.1}\text{Co}_{0.45}\text{V}_{0.45}\text{Al}$ with $L2_1$ structure.

band gap (≈ 380 meV) in one of the spin channels indicates half-metallic ferrimagnetism.

IV. SUMMARY AND CONCLUSION

A common misconception about Heusler alloys arises from the misinterpretation of electron-counting rules. For example, the prototype Cu_2MnAl , which has 32 valence electrons, exhibits a magnetic moment of approximately $3.7 \mu_B$ per primitive cell, which means it does not follow the proposed rules. Based on these rules, it is often claimed that Heusler alloys with $n_v = 24$ carry no magnetic moments and do not exhibit magnetic order. However, this assumption is incorrect, as demonstrated by the present work and earlier studies. Therefore, these rules have limited validity, especially for half-metallic ferromagnets. The original Slater–Pauling concept [16,54–56] and Néel’s theory of ferrimagnets [1] extend well beyond such simplistic rules, even though such rules might yield rough estimates of the magnitudes of magnetic moments.

In the present work, the thermomagnetic properties and electronic structures of Heusler alloys were investigated by first-principles calculations, following the design scheme for half-metallic completely compensated ferrimagnets. The results indicate that these alloys belong to the same class as Néel’s L -type ferrimagnets, a class of materials that has been largely overlooked in the literature.

For two sublattice alloys with an $L2_1$ structure and 24 valence electrons, the total magnetization vanishes at $T = 0$ but remains finite up to the Curie temperature. Similar behavior is observed for most three-sublattice alloys with a Y -type structure. In slightly off-stoichiometric alloys, the compensation point shifts to temperatures above 0 K. This occurs in alloys with fewer than 24 valence electrons, whereas the compensation point disappears for $n_v > 24$. All Heusler alloys investigated in this study exhibit half-metallic character. The band gap in the minority states remains unchanged, with chemical disorder scattering exerting strong influence only in the metallic majority states. Thus, alloys with random site occupations are expected to show higher resistivity and less pronounced temperature dependence compared with well-ordered compounds with 2:1:1 stoichiometry.

Lastly, as demonstrated for $\text{Mn}_{2.1}\text{Co}_{0.45}\text{V}_{0.45}\text{Al}$, multiple compensation points can exist in Heusler alloys. In addition to the compensation at 0 K, a second compensation point is found below the Curie temperature. The low residual magnetization of this material makes it suitable for spintronic applications. Overall, the results demonstrate that Heusler alloys can be designed to have multiple compensation points and a magnetization approaching nearly zero up to the Curie point.

ACKNOWLEDGMENTS

We are grateful to Professor J. Kübler (now deceased) for valuable suggestions and insights and for establishing the theoretical basis for describing Mn-based Heusler compounds as completely compensated ferrimagnets. This work was supported by the European Research Council (ERC) Advanced

Grant “Strain-Free All Heusler Alloy Junctions SAHAJ”; Grant agreement ID 101097475.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX: NÉEL’S THEORY OF FERRIMAGNETS REVISITED

Néel’s work on ferrimagnets and antiferromagnets is based on the molecular field theory. The first part of Ref. [1] focuses on the temperature dependence of the magnetization of ferrites. Therefore, some parameters cannot be used unchanged for more general cases. However, this problem only affects the parameter space, not the different shapes of the magnetization curves. The second part of Ref. [1], on particular systems, provides hints about how other parameter sets influence the magnetization of ferrimagnets.

The starting point is a material with two magnetic sublattices, A and B. The relative fraction λ of ions occupies sublattice A, while the relative fraction μ occupies sublattice B. The important side condition is $\lambda + \mu = 1$. According to Néel, the molecular fields acting on the sublattices of this material can be written as follows:

$$\begin{aligned} h_A &= n(\lambda\alpha M_A + \epsilon\mu M_B), \\ h_B &= n(\epsilon\lambda M_A + \mu\beta M_B), \end{aligned} \quad (\text{A1})$$

where M_A and M_B are the sublattice magnetizations. The intrasublattice molecular field coefficients are A_{AA} and A_{BB} , and the intersublattice coefficient is $A_{AB} = A_{BA}$. The relative interactions are given by $\alpha = A_{AA}/A_{AB}$ and $\beta = A_{BB}/A_{AB}$. The relationship between the molecular field coefficients and exchange integrals is discussed in Ref. [1]. The sign of the intersublattice molecular field coefficient is given by $\epsilon = A_{AB}/|A_{AB}|$, and its magnitude is $n = |A_{AB}|$. A positive

interaction ($\epsilon = +1$) favors an antiparallel orientation of the sublattice magnetizations, resulting in ferromagnetic order. Conversely, a negative interaction ($\epsilon = -1$) favors antiparallel alignment. Depending on the sublattice occupation, this results in ferrimagnetic or antiferromagnetic order. The temperature-dependent magnetization of the sublattices is described by the following set of coupled equations:

$$\begin{aligned} M_A &= m_A B_{j_A}(z_A), \\ M_B &= m_B B_{j_B}(z_B), \\ z_A &= \frac{m_A \cdot h_A}{k_B \cdot T}, \\ z_B &= \frac{m_B \cdot h_B}{k_B \cdot T}, \end{aligned} \quad (\text{A2})$$

where $B_j(z)$ is the Brillouin function with the Langevin variable z including the Boltzmann constant k_B . The sublattice magnetizations at $T = 0$ are represented by m_A and m_B . The total magnetization is given by $M(T) = \lambda M_A(T) + \mu M_B(T)$. This value is determined by solving the aforementioned set of coupled equations. Equations (A1) and (A2) together form the magnetic equation of state of the two-sublattice system.

The following restrictions appear in Ref. [1], necessitating changes for the Heusler compounds: The special ratio of sites $\lambda/\mu = 2 : 3$ is used for ferrites, whereas its value is $\lambda/\mu = 1 : 2$ in regular Heusler compounds with the $L2_1$ structure. Additionally, trivalent Fe ions with the same magnetic moments $m_A = m_B = m$ at $T = 0$ are assumed. However, the sublattice magnetizations differ in the case of Heusler compounds. Completely compensated ferrimagnets, in particular, require a ratio of $m_A/m_B = \mu/\lambda = 2 : 1$ owing to the site ratio of $1 : 2$. The resulting magnetic equation of state diagram is shown in Fig. 14. It distinguishes regions of different types of magnetism as a function of the intersublattice molecular field coefficients α and β . No magnetic order is possible in the region below the line $\overline{ASB}$. The line $\overline{ASB}$ is defined by $\alpha \cdot \beta = 1$ ($\alpha, \beta < 0$). In this region, the material is paramagnetic.

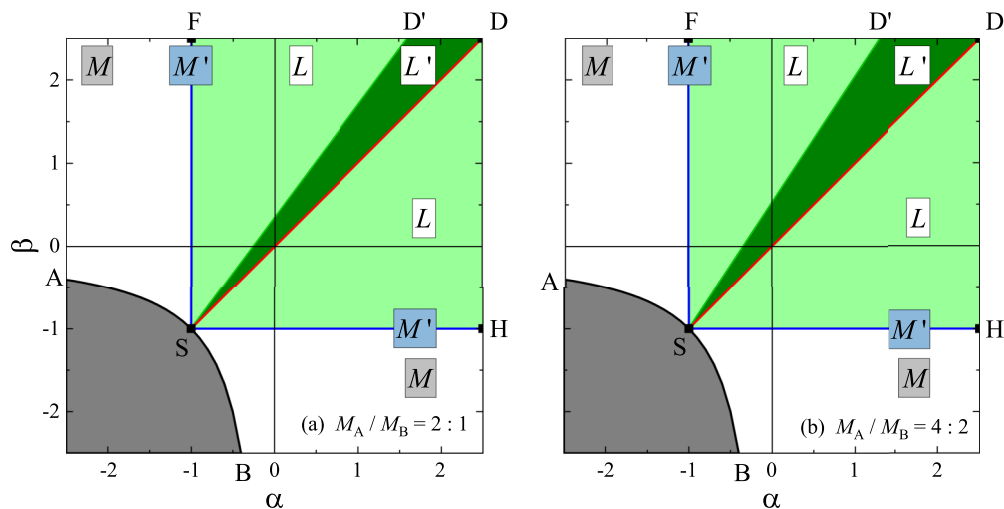


FIG. 14. Magnetic equation of state diagram for compensated ferrimagnetic Heusler compounds ($\epsilon = -1$). The diagram is plotted for two different ratios of the initial magnetic moments, (a) 2:1 and (b) 4:2. The site ratio of regular Heusler compounds with $L2_1$ structure is $\lambda/\mu = 1 : 2$, considering the multiplicity of the $4b$ and $8c$ Wyckoff positions.

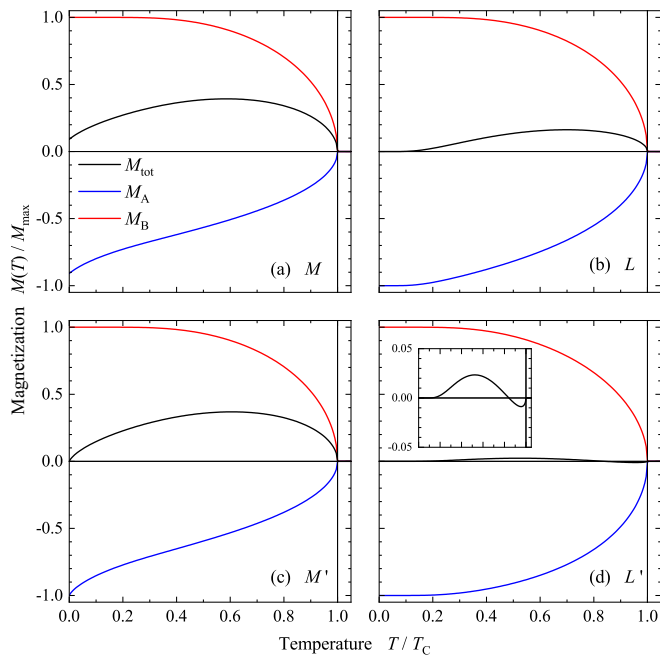


FIG. 15. Magnetization types for compensated ferrimagnetic Heusler compounds ($\epsilon = -1$). The types are plotted for an initial magnetic moment ratio of 4:2 and $\lambda/\mu = 1 : 2$.

Four types of magnetization curves appear in the equation of state diagram for completely compensated Heusler ferrimagnets, i.e., L and L' , as well as M and M' . At the line $\overline{SD}$, where $\alpha = \beta$, the sign of the sublattice magnetizations changes. Exchanging the values of λ and μ ($\lambda : \mu \rightarrow \mu : \lambda$) merely results in an exchange of the α and β axes in the diagram. M' appears only along the lines $\overline{SF}$ and $\overline{SH}$ where one of the coefficients is -1 . The region inside the line $\overline{SDD'S}$, in which L' appears, depends on the ratio of the initial magnetic moments at $T = 0$. The same is true for the shapes of the curves, which are influenced by the Brillouin functions that depend on the magnitude of the magnetic moments as well. Figure 15 shows the different types of temperature-dependent magnetization curves.

Néel did not explicitly mention the two primed temperature dependencies. M' has the same shape as M in Ref. [1]. However, the magnetization is zero at $T = 0$. M does not exhibit a compensation point below the Curie temperature; it is plotted for completeness. Finally, L' differs from L in that it has an additional zero-crossing in its temperature dependence. The total magnetization remains small for the L' type. The exact position of the additional zero-crossing depends on the reduced molecular field coefficients α and β .

- [1] L. Néel, Magnetic properties of ferrites: Ferrimagnetism and antiferromagnetism, *Ann. Phys.* **12**, 137 (1948).
- [2] L. Néel, Magnetic properties of ferrites: Ferrimagnetism and antiferromagnetism, in *Selected Works of Louis Néel*, edited by N. Kurti (CRC Press, Boca Roton, London, New York, 1988), Chap. 2, p. 63.
- [3] L. Néel, Magnetism and local molecular field, *Science* **174**, 985 (1971).
- [4] J. S. Smart, The Néel theory of ferrimagnetism, *Am J. Phys.* **23**, 356 (1955).
- [5] B. Barbara, Louis Néel: His multifaceted seminal work in magnetism, *C. R. Phys.* **20**, 631 (2019).
- [6] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/1913-4x5y> for more details on symmetry considerations for ferrimagnetic and antiferromagnetic materials, Néel's diagrams and types, thermomagnetic properties of selected Heusler compounds, relaxed lattice parameters, and proposed compensated ferrimagnetic Heusler alloys.
- [7] Fr. Heusler, Über Magnetisierbare Manganlegierungen, *Verh. d. DPG* **5**, 219 (1903).
- [8] C. Felser and G. H. Fecher, *Spintronics: From Materials to Devices* (Springer Verlag, Dordrecht, 2013).
- [9] C. Felser and A. Hirohata, Heusler alloys: Properties, growth, applications, in *Springer Series in Materials Science* (Springer Verlag, Cham, 2016), Vol. 222.
- [10] R. A. de Groot, Half-metallic magnetism in the 1990s, *Phys. B* **172**, 45 (1991).
- [11] H. van Leuken and R. A. de Groot, Half-metallic antiferromagnets, *Phys. Rev. Lett.* **74**, 1171 (1995).
- [12] S. Wurmehl, H. C. Kandpal, G. H. Fecher, and C. Felser, Valence electron rules for prediction of half-metallic compensated-ferrimagnetic of Heusler compounds with complete spin polarization, *J. Phys.: Condens. Matter* **18**, 6171 (2006).
- [13] J. Kübler, A. R. William, and C. B. Sommers, Formation and coupling of magnetic moments in Heusler alloys, *Phys. Rev. B* **28**, 1745 (1983).
- [14] R. Stinshoff, G. H. Fecher, S. Chadov, A. K. Nayak, B. Balke, S. Ouardi, T. Nakamura, and C. Felser, Half-metallic compensated ferrimagnetism with a tunable compensation point over a wide temperature range in the Mn-Fe-V-Al Heusler system, *AIP Adv.* **7**, 105009 (2017).
- [15] R. Stinshoff, A. K. Nayak, G. H. Fecher, B. Balke, S. Ouardi, Y. Skourski, T. Nakamura, and C. Felser, Completely compensated ferrimagnetism and sublattice spin crossing in the half-metallic Heusler compound $\text{Mn}_{1.5}\text{FeV}_{0.5}\text{Al}$, *Phys. Rev. B* **95**, 060410(R) (2017).
- [16] G. H. Fecher, H. C. Kandpal, S. Wurmehl, C. Felser, and G. Schönhense, Slater-Pauling rule and Curie-temperature of Co_2 -based Heusler compounds, *J. Appl. Phys.* **99**, 08J106 (2006).
- [17] J. Kübler, G. H. Fecher, and C. Felser, Understanding the trend in the Curie temperatures of Co_2 -based Heusler compounds: *Ab initio* calculations, *Phys. Rev. B* **76**, 024414 (2007).
- [18] M. Gibertini, M. Koperski, A. F. Morpurgo, and K. S. Novoselov, Magnetic 2D materials and heterostructures, *Nat. Nanotechnol.* **14**, 408 (2019).
- [19] X. Jiang, Q. Liu, J. Xing, N. Liu, Y. Guo, Z. Liu, and J. Zhao, Recent progress on 2D magnets: Fundamental mechanism, structural design and modification, *Appl. Phys. Rev.* **8**, 031305 (2021).
- [20] Y. Liu, S.-D. Guo, Y. Li, and C.-C. Liu, Two-dimensional fully compensated ferrimagnetism, *Phys. Rev. Lett.* **134**, 116703 (2025).

- [21] S.-D. Guo, J. He, and Y. S. Ang, Achieving fully compensated ferrimagnetism through two-dimensional CrI₃/CrGeTe₃ heterojunctions, *Appl. Phys. Lett.* **127**, 232401 (2025).
- [22] Y. Li, D. Zheng, C. Liu, C. Zhang, B. Fang, A. Chen, Y. Ma, A. Manchon, and X. Zhang, Current-induced magnetization switching across a nearly room-temperature compensation point in an insulating compensated ferrimagnet, *ACS Nano* **16**, 8181 (2022).
- [23] R. A. de Groot and K. H. J. Buschow, Recent developments in half-metallic magnetism, *J. Magn. Magn. Mater.* **54-57**, 1377 (1986).
- [24] T. Tsuchiya, J. Okabayashi, and M. Mizukami, Element-specific magnetic properties of compensated ferrimagnet Mn₂Co_{1-x}V_xAl Heusler alloy films, *J. Magn. Magn. Mater.* **540**, 168437 (2021).
- [25] H. Ebert, Fully relativistic band structure calculations for magnetic solids—formalism and application, in *Electronic Structure and Physical Properties of Solids*, edited by H. Dreyssé, Lecture Notes in Physics, Vol. 535 (Springer, Berlin, Heidelberg, 1999).
- [26] H. Ebert, The Munich SPR-KKR package, Version 8.6 (2005), <https://olymp.cup.uni-muenchen.de/index.php/de/software>.
- [27] H. Ebert, D. Ködderitzsch, and J. Minar, Calculating condensed matter properties using the KKR-Green's function method—Recent developments and applications, *Rep. Prog. Phys.* **74**, 096501 (2011).
- [28] P. Soven, Coherent-potential model of substitutional disordered alloys, *Phys. Rev.* **156**, 809 (1967).
- [29] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized gradient approximation made simple, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [30] J. S. Faulkner and G. M. Stocks, Calculating properties with the coherent-potential approximation, *Phys. Rev. B* **21**, 3222 (1980).
- [31] A. I. Liechtenstein, M. I. Katsnelson, V. P. Antropov, and V. A. Gubanov, Local spin density functional approach to the theory of exchange interactions in ferromagnetic metals and alloys, *J. Magn. Magn. Mater.* **67**, 65 (1987).
- [32] P. W. Anderson, Theory of magnetic exchange interactions: Exchange in insulators and semiconductors, *Solid State Phys.* **14**, 99 (1963).
- [33] H. Ebert and S. Mankovsky, Anisotropic exchange coupling in diluted magnetic semiconductors: *Ab initio* spin-density functional theory, *Phys. Rev. B* **79**, 045209 (2009).
- [34] S. Mankovsky and H. Ebert, Accurate scheme to calculate the interatomic Dzyaloshinskii-Moriya interaction parameters, *Phys. Rev. B* **96**, 104416 (2017).
- [35] S. Mankovsky and H. Ebert, First-principles calculation of the parameters used by atomistic magnetic simulations, *Electron. Struct.* **4**, 034004 (2022).
- [36] R. F. L. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. A. Ellis, and R. W. Chantrell, Atomistic spin model simulations of magnetic nanomaterials, *J. Phys.: Condens. Matter* **26**, 103202 (2014).
- [37] R. F. L. Evans, U. Atxitia, and R. W. Chantrell, Quantitative simulation of temperature-dependent magnetization dynamics and equilibrium properties of elemental ferromagnets, *Phys. Rev. B* **91**, 144425 (2015).
- [38] J. Hopkinson, XIV. Magnetic and other physical properties of iron at a high temperature, *Philos. Trans. A Math. Phys. Eng. Sci.* **180**, 443 (1889).
- [39] G. L. Ferreira-Fraga, L. A. Borba, and P. Pureur, Impedance and initial magnetic permeability of the Heusler compounds Pd₂MnSn and Pd₂MnSb near the Curie temperature, *Phys. Rev. B* **74**, 064427 (2006).
- [40] P. Blaha, K. Schwarz, F. Tran, R. Laskowski, G. K. H. Madsen, and L. D. Marks, WIEN2k: An APW+lo program for calculating the properties of solids, *J. Chem. Phys.* **152**, 074101 (2020).
- [41] S. J. Joshua, *Symmetry Principles and Magnetic Symmetry in Solid State Physics* (Adam Hilger, IOP Publishing Ltd., Bristol, 1991).
- [42] B. J. Campbell, H. T. Stokes, J. M. Perez-Mato, and J. Rodriguez-Carvajal, Introducing a unified magnetic space-group symbol, *Acta Cryst. A* **78**, 99 (2022).
- [43] H. T. Stokes and D. M. Hatch, Program for identifying the space group symmetry of a crystal, *J. Appl. Cryst.* **38**, 237 (2005).
- [44] V. Niculescu, T. J. Burch, K. Raj, and J. I. Budnick, Properties of Heusler-type materials Fe₂TsI and FeCo₂Si, *J. Magn. Magn. Mater.* **5**, 60 (1977).
- [45] K. H. J. Buschow and P. G. van Engen, Magnetic and magneto-optical properties of Heusler alloys based on aluminium and gallium, *J. Magn. Magn. Mater.* **25**, 90 (1981).
- [46] V. Y. Markiv, E. I. Gladyshevskii, and Y. B. Kuzma, New ternary compounds with a structure of the MnCu₂Al type, *Dopovidi Akademii Nauk Ukrain's'koi RSR* **10**, 1329 (1962).
- [47] L. Basit, G. H. Fecher, S. Chadov, B. Balke, and C. Felser, Quaternary Heusler compounds without inversion symmetry: CoFe_{1+x}Ti_{1-x}Al and CoMn_{1+x}V_{1-x}Al, *Eur. J. Inorg. Chem.* **2011**, 3950 (2011).
- [48] I. Galanakis and E. Sasioglu, High *T_C* half-metallic fully-compensated ferrimagnetic Heusler compounds, *Appl. Phys. Lett.* **99**, 052509 (2011).
- [49] T. Graf, F. Casper, J. Winterlik, B. Balke, G. H. Fecher, and C. Felser, Crystal structure of new Heusler compounds, *Z. Anorg. Allg. Chem.* **635**, 976 (2009).
- [50] R. Gavrea, R. Hirian, O. Isnard, V. Pop, and D. Benea, Investigations on compensated ferrimagnetism in the Mn₂Co_{0.5}V_{0.5}Al Heusler alloy, *Solid State Commun.* **309**, 113812 (2020).
- [51] M. A. Seregina, D. Yu. Karpenkov, E. A. Kolesnikov, M. V. Gorshenkov, A. Yu. Degtyarenko, S. V. Taskaev, P. N. Degtyarenko, X. Xu, and V. V. Khovaylo, Compensated ferrimagnetism and compensation temperatures in Mn_{2-2x}Co_{0.5+x}V_{0.5+x}Ga Heusler alloys, *J. Magn. Magn. Mater.* **562**, 169808 (2022).
- [52] S. I. Ohkoshi, Y. Abe, A. Fujishima, and K. Hashimoto, Design and preparation of a novel magnet exhibiting two compensation temperatures based on molecular field theory, *Phys. Rev. Lett.* **82**, 1285 (1999).
- [53] X. H. Chen, K. Q. Wang, P. H. Hor, Y. Y. Xue, and C. W. Chu, Anomalies at the compensation temperature in the zero-magnetization ferromagnet (Sm, Gd)Al₂, *Phys. Rev. B* **72**, 054436 (2005).
- [54] J. C. Slater, The ferromagnetism of nickel. II. Temperature effects, *Phys. Rev.* **49**, 931 (1936).
- [55] L. Pauling, The nature of the interatomic forces in metals, *Phys. Rev.* **54**, 899 (1938).
- [56] J. Kübler, First principle theory of metallic magnetism, *Phys. B* **127**, 257 (1984).