

Half-Metallic Full-Heusler and Half-Heusler Compounds with Perpendicular Magnetic Anisotropy

Sergey V. Faleev,* Panagiotis Ch. Filippou, Chirag Garg, Jaewoo Jeong, Mahesh G. Samant, and Stuart S. P. Parkin*

Herein, a mechanism is proposed as to how thin films formed from a Heusler compound can simultaneously have both perpendicular magnetic anisotropy (PMA) and be half-metallic. It is proposed that a thin film formed from a half-metallic full-Heusler or half-Heusler compound that is cubic in the bulk can undergo a tetragonal distortion by adopting the lattice constant of the underlayer material during the thin film deposition process. The value of this distortion can be tuned by using underlayer materials with different in-plane lattice constants. The distortion can thereby be optimized so that it is large enough to give rise to sufficient PMA, while, simultaneously, small enough to retain the half-metallic properties (and, therefore, high tunneling magnetoresistance properties) of the Heusler compound. Density functional theory (DFT) calculations that are carried out for 90 full-Heuslers and 147 half-Heuslers that have been identified in the literature as half-metals show that of these, 14 full-Heusler and 59 half-Heusler compounds display both half-metallicity and PMA for optimal tetragonal distortions.

1. Introduction

Spin-transfer torque magnetic random-access memory (STT-MRAM) is considered today as the most promising contender for a "universal memory" that combines all the strengths and none of the weaknesses of existing memory types.^[1] STT-MRAM combines nonvolatility, high read and write speeds, unlimited endurance, reconfigurable logic functions, and

S. V. Faleev, P. C. Filippou, C. Garg, M. G. Samant, S. S. P. Parkin IBM Research – Almaden San Jose, CA 95120, USA E-mail: sergey.faleev@ibm.com

J. Jeong Samsung Electronics San Jose, CA 95124, USA

S. S. P. Parkin Max Plank Institute for Microstructure Physics Weinberg 2, 06120 Halle (Saale), Germany E-mail: stuart.parkin@mpi-halle.mpg.de

The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/pssb.202200531.

© 2023 The Authors. physica status solidi (b) basic solid state physics published by Wiley-VCH GmbH. This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

DOI: 10.1002/pssb.202200531

ultralow power consumption. Key to the successful development of this technology is the discovery of new magnetic materials for the magnetic tunnel junction (MTJ) memory elements that have sufficient stability against thermal fluctuations to sustain deeply scaled devices.

The magnetic electrodes that form the MTJ must possess sufficient perpendicular magnetic anisotropy (PMA) that their magnetizations lie perpendicular to the plane of the MTJ device because this allows for lower currents to switch the magnetization of the electrode that forms the memory layer of the device using spin torque.^[2,3] The most promising magnetic materials to date are considered to be alloys formed from Co, Fe, and B, in conjunction with MgO tunnel barriers.^[3–5] Unfortunately, the PMA of the Co–Fe–B layers arises from

the interfaces between these layers and the tunnel barrier and/or under- or overlayers and is too weak to overcome thermal fluctuations when the diameter of the MTJ has a critical size of less than \approx 20 nm to allow for the required 10 year memory stability.

Magnetic materials in which the PMA is derived from volume magnetocrystalline anisotropy (MCA) are thus needed to allow for increased PMA values, robust to scaling. Two of the most promising classes of such materials are the full-Heusler compounds that have the chemical formula X₂YZ and half-Heusler compounds that have the chemical formula XYZ, wherein X and Y are transition metals or lanthanides (rare-earth metals), and Z is the main group element.^[6] In the case of half-Heuslers, X and Y can also be alkali or alkaline earth metals. While the parent Heusler compounds are cubic and do not exhibit significant magnetic anisotropy, the structure of some of these compounds is found to be tetragonally distorted and thus can potentially display large PMA. (In this article, we will use the term "Heuslers" as a common name for both full-Heuslers and half-Heuslers.)

Some examples of tetragonal Heusler compounds are $Mn_{3-x}Ga^{[7]}$ and Mn_3Ge .^[8] Thin films of these materials have been shown to exhibit large PMA for films grown epitaxially on single-crystalline substrates such as STiO₃(001) or MgO(001),^[8-12] as well as on amorphous substrates formed from Si(001)/SiO₂.^[13,14] Unfortunately, the experimental values of the tunneling magnetoresistance (TMR) for MTJs with $Mn_{3-x}Ga$ or Mn_3Ge electrodes and MgO tunneling barriers were found to be very small, far below the values needed for MRAM applications.^[13] Tetragonal Heuslers with theoretically predicted large

PMA that could potentially show high TMR ether due to the large spin polarization or due to so-called Brillouin zone spin filtering effect have been recently suggested in ref. [15]. While tetragonal ground state and high PMA have been experimentally verified for some of suggested compounds,^[15] there is no experimental conformation yet of high TMR in MTJs with these electrodes.

ADVANCED SCIENCE NEWS _____ www.advancedsciencenews.com

On the other hand, very high TMR values have been reported for MTJs with electrodes formed from cubic full-Heuslers, as high as 354% at room temperature (RT) for Co₂MnSi.^[16] Such a high TMR is attributed to the half-metallicity of these compounds.^[16,17] Unfortunately, all half-metallic full-Heusler and half-Heusler compounds known to date are cubic, and thus have negligible bulk magnetocrystalline anisotropy.

In order to have both PMA and half-metallicity, we propose to create a tetragonal distortion in thin films formed from nominally cubic, half-metallic Heuslers by preparing these films on underlayer materials that have corresponding (smaller or larger) in-plane lattice constants. The distortion should be large enough to make the Heusler compound tetragonal with sufficient PMA, while, simultaneously, small enough to retain the half-metallic properties and, therefore, high TMR of the compound. In this article, we systematically study the properties (PMA and halfmetallicity) of Heusler compounds that have been identified in the literature as being half-metallic, as a function of their in-plane lattice constant that we vary in the vicinity of the equilibrium cubic lattice constant.

2. Half-Metallic and PMA Properties of Full-Heusler Compounds

2.1. Crystal Structure

Cubic full-Heusler compounds X_2YZ can have a regular structure or an inverse structure. These two crystal structures are shown schematically in **Figure 1**a,c. Four sites form four *fcc* sublattices: site Z (occupied by atom Z); site II, octahedrally coordinated by Z; and two equivalent sites I that are tetrahedrally coordinated by Z. In the regular Heusler structure shown in Figure 1a, two X atoms [red, labeled as X(I)] have an identical environment—they are located on sites I in the same *xy*-plane. In this structure, the Y atom (cyan) on site II and Z atom (grey) are located in a second *xy*-plane. In the inverse Heusler structure shown in Figure 1c, two X atoms have different environments—one X atom [red, labeled as X(I)] is located on site I in one *xy*-plane together with a Y atom (cyan), while another X atom [orange, labeled as X(II)] is located on site II in a second *xy*-plane with the Z atom (gray).

Regular (Figure 1b) and inverse (Figure 1d) tetragonal Heusler structures can be obtained from regular and inverse cubic structures, correspondingly, by stretching (or compressing) the parent cubic structure along the z-axis (note that the z-axis is along the film normal). Tetragonal unit cells shown in Figure 1b,d are rotated by 45° around the z-axis relative to the parent cubic structures shown in Figure 1a,c, respectively. (Note that only a subset of the atoms from Figure 1a,c are shown in Figure 1b,d.) The lattice constant a_{cub} of the cubic Heusler is indicated in Figure 1a and the lattice constants a and c of the tetragonal Heusler are shown in Figure 1b. For characterization of the tetragonal unit cell we use the dimensionless parameter c' = c/(2a) that is equal to $1/\sqrt{2} \approx 0.707$ for the cubic structure, and varies between 0.6 and 0.9 for most of the tetragonal Heuslers we consider here (see Table 3, 6, 7). Note that for c' = 1 the tetragonal structure of a full-Heusler becomes the fcc structure if all four atoms of the compound are considered as equivalent.

2.2. Computational Details

We performed DFT calculations for both the regular and inverse structures with various initial magnetic configurations (four possible relative magnetic orientations of X. X. and Y atoms) and various cubic lattice constants for 90 cubic full-Heusler compounds that have been identified as half-metals in the literature. For selected compounds (see Table 3), we also performed calculations for tetragonally distorted structures. For the DFT calculations, we used the generalized gradient approximation of the density functional theory (GGA/DFT) implemented within the VASP program^[18] with projector augmented wave potentials^[19,20] and the PBE GGA/DFT functional.^[21] The convergence of the results presented below was verified for selected compounds by varying the number of divisions in reciprocal space from $10 \times 10 \times 8$ to $18 \times 18 \times 14$ and by varying the energy cutoff that defines the plane waves basis in VASP code from 400 to 520 eV. The final density of states (DOS) calculations for all compounds were performed with $12 \times 12 \times 12$ divisions in reciprocal space and 520 eV energy cutoff that provided better than 0.01 eV accuracy for the bandgap values. Structure optimization and DOS calculations were performed without inclusion of the spin-orbit interaction.



Figure 1. a) Regular and c) inverse cubic Heusler structure. b) Regular and d) inverse tetragonal Heusler structure.



The MCA energy per formula unit, $K_{\rm mc}$, of the tetragonally distorted Heusler compounds is calculated as the difference between the total energies of the magnetic states with magnetization directed along the *x*-axis and the *z*-axis, i.e., $K_{\rm mc} = E(100) - E(001)$, where positive $K_{\rm mc}$ corresponds to out-of-plane magnetization. For MCA calculations, we used finer $20 \times 20 \times 20$ mesh in reciprocal space and included spin–orbit interactions self-consistently in the DFT cycle. We also calculated the volume magnetic anisotropy, $K_{\rm v} = K_{\rm mc} - K_{\rm sh}$, where $K_{\rm sh} = \mu_0 M_{\rm s}^2 V/2$ is the shape anisotropy energy of a thin film per unit cell volume *V*, $M_{\rm s}$ is the saturation magnetization, and μ_0 is the vacuum permeability.

The Curie temperature, $T_{\rm C}$, was calculated within the standard mean-field approximation (MFA)^[22] using the exchange constants, J_{ij} , of the effective Heisenberg Hamiltonian (*i* and *j* are the site indexes). In this approach, $T_{\rm C}$ can be estimated as^[23] $k_{\rm B} T_{\rm C} = 2/3J_{\rm max}$, where $J_{\rm max}$ is the maximal eigenvalue of the (4 × 4) $J^{\mu\nu}$ matrix, with $J^{\mu\nu} = \sum_{j \in \nu} J_{0j}$. Here, 0 is fixed index for the sublattice μ and the sum is taken over sites in the sublattice ν . The exchange constants J_{ij} were calculated with the QUESTAAL code (www.questaal.org) using a Green's function approach implemented within the LMTO-ASA framework.^[24,25]

2.3. Results and Discussion

The results of calculations for full-Heuslers compounds are summarized in Table 1-3. We found that out of the 90 considered full-Heusler cubic compounds, 35 compounds are either half-metals or near half-metals (these compounds are included in Table 1) and 55 compounds are neither half-metals nor near half-metals (these are compounds included in Table 2). Here, we define a compound as being a half-metal if it has a bandgap in one of the spin channels and the Fermi energy is located within this bandgap (lower edge of the bandgap, E_{\min} , is located below the Fermi energy, $E_{\rm F}$, while the upper edge of the bandgap, $E_{\rm max}$, is located above the Fermi energy). We define a compound as being a near-half-metal if it has a bandgap in one of the spin channels and either E_{\min} is located not too far above $E_{\rm F}$ such that $0 < E_{\min} - E_F \le 0.1 \text{ eV}$, or E_{\max} is located not too far below E_F so that $0 < E_{\rm F} - E_{\rm max} \leq 0.1$ eV. Calculated values of $E_{\rm min}$, $E_{\rm max}$ (where we set $E_{\rm F} = 0$), and the bandgap, $E_{\rm gap} = E_{\rm max} - E_{\rm min}$, are presented in Table 1. The next-to-last column of Table 1 also indicates if a compound is a half-metal (HM) or a near-half-metal (near-HM). The bandgaps of all compounds presented in Table 1 are in the minority spin channel, except for Mn₂VAl and Cr₂MnAl where the bandgap is in the majority spin channel (which corresponds to a negative magnetic moment m (in μ_B) for these compounds according to the Slater–Pauling rule,^[26–28] m = N - 24, where N is the total number of valence electrons per formula unit).

Table 1 and 2 show the calculated values of the cubic lattice constants in the regular structure, a_r , and inverse structure, a_i , the total magnetic moment in the regular structure, m_r , and the inverse structure, m_i , the difference between the total energy of the regular and inverse structures, $E_r - E_i$, and an indicator if the ground state is a regular or inverse structure (column "ground state"). The ground state has an inverse structure if $E_r - E_i > 0$ and a regular structure if $E_r - E_i < 0$. The values of E_{max} , E_{min} , E_{gap} , and the HM/near-HM indicator that are shown in Table 1 correspond to the ground state of the compound.

References to published papers where a specific compound was identified as being a half-metal are presented in Table 1 and 2. We have found that compounds presented in Table 2 are not half-metallic in the ground state, but many compounds in Table 2 that have the regular structure ground state are half-metallic in the inverse structure (this can be seen from the nonfractional values of m_i in Table 2, which are in agreement with the Slater-Pauling rule). Some of the compounds in Table 2 have a bandgap in one spin channel, but either with E_{\min} located too far above $E_{\rm F}$, with $E_{\rm min} - E_{\rm F} > 0.1 \, {\rm eV}$, or $E_{\rm max}$ located too far below $E_{\rm F}$, with $E_{\rm F} - E_{\rm max} > 0.1$ eV. Also, some of the compounds in Table 2 do not have a bandgap but have a very small density of states (DOS) in one of the spin channels at or near $E_{\rm F}$. We attribute different conclusions regarding the half-metallicity of compounds presented in Table 2 obtained in the present work and in the literature to the sensitivity of the bandgap (or even existence of the bandgap) of these compounds to the details of the computational method used (e.g., the chosen DFT functional). Also, the authors of ref. [29] only considered inverse structures, while we found that for many of the Heuslers considered in ref. [29] the ground state is the regular structure.

Table 3 presents results of calculations of the PMA and halfmetallic properties of 22 half-metals from Table 1 when these compounds undergo a tetragonal distortion. The list of compounds in Table 3 is that of Table 1 with exclusion of 11 near-half-metals and two half-metallic compounds, Cr₂MnAl and Cr₂NiAl, whose bandgap, $E_{\rm gap} \leq 0.05$ eV, is too small. As mentioned in the introduction, the tetragonal distortion of a thin film of Heusler compound is expected to be a result of the Heusler compound adopting (via thin film epitaxy) the lattice constant of the underlayer material (that could be larger or smaller than the cubic lattice constant of the Heusler) during the thin film deposition process.

Table 3 shows the indicator of the ground state structure (regular or inverse), the lattice constant, a_c , the total magnetic moment, $m_{\rm c}$, the bandgap, $E_{\rm gap}$, and the Curie temperature, $T_{\rm C}$, calculated for the ground state cubic structure. (Note that the lattice constant a_c corresponds to a shown in Figure 1b and is related to $a_{\rm cub}$ shown in Figure 1a as $a_{\rm c} = a_{\rm cub}/\sqrt{2}$). Table 3 also shows the minimum, a_{\min} , and the maximum, a_{\max} , values of the in-plane lattice constant for which the tetragonally distorted compound retains its half-metallicity. The width of the half-metallic range, $w_{\rm hm}$, defined as $w_{\rm hm} = a_{\rm max} - a_{\rm min}$ serves as a measure of the robustness of the half-metallic properties of the compound. Larger values of $w_{\rm hm}$ and larger values of the bandgap, E_{gap} , result in more stable half-metallic properties (stable, nonvanishing bandgap less susceptible to details of calculations or other effects). Effects that can affect half-metallic properties include change in the in-plane and out-of-plane lattice constants, details of the computational method (the chosen DFT functional, inclusion of the spin-orbit coupling, use of beyond DFT methods, such as LDA + U, GGA + U, or GW), finite temperature effects, effects of surfaces and interfaces, and effects of defects such as impurities or dislocations.

The a_{opt} shown in Table 3, are the in-plane lattice constants that provide the largest (optimal) MCA energy, K_{mc} , for *a* in the half-metallicity range ($a_{\min} \leq a_{\text{opt}} \leq a_{\max}$). The values, c'_{opt} are the out-of-plane dimensionless constants that correspond to a_{opt} .



Table 1. Calculated cubic lattice constant of full-Heuslers that we have identified as half-metals or near-half-metals (see definition of near-half-metals in the text) in the regular structure, a_r , and in the inverse structure, a_i , total magnetic moment in the regular structure, m_r , and in the inverse structure, a_i , total magnetic moment in the regular structure, m_r , and in the inverse structure, m_i , the difference between the total energy in the regular and inverse structures, $E_r - E_i$, bottom edge of the bandgap, E_{min} (where we set $E_F = 0$), top edge of the bandgap, E_{max} , and the value of the bandgap, $E_{gap} = E_{max} - E_{min}$ in the ground state (as shown in the "Ground state" column).

	a _r [Å]	т, [µ _В]	<i>a</i> _i [Å]	т _і [µ _В]	$E_r - E_i$ [eV]	Ground state	E _{min} [eV]	E _{max} [eV]	Egap [eV]		
V ₂ MnAl	4.22	1.422	4.20	2.000	0.266	Inv	-0.07	0.01	0.08	НМ	[29]
Cr ₂ MnAl	4.18	1.997	4.11	2.000	0.371	Inv	-0.01	0.03	0.04	НМ	[29]
Cr ₂ NiAl	4.24	6.658	4.12	1.000	1.025	Inv	-0.04	0.01	0.05	НМ	[29]
Cr ₂ CoSi	4.03	2.829	4.00	1.001	0.765	Inv	0.09	0.11	0.02	Near-HM	[29]
Cr₂ZnSi	4.20	5.130	4.13	0.000	0.007	Inv	-0.15	0.28	0.43	НМ	[29]
Mn₃Al	4.09	0.000					-0.09	0.42	0.51	НМ	[29]
Mn ₂ FeAl	4.11	7.010	4.06	1.000	0.332	Inv	0.05	0.49	0.44	Near-HM	[29]
Mn₃Si	3.99	1.001					0.10	0.49	0.39	Near-HM	[29]
Mn_2FeSi	3.96	2.062	3.96	2.001	0.388	Inv	0.09	0.33	0.24	Near-HM	[29]
Mn₃As	4.04	2.000					0.06	0.25	0.19	Near-HM	[29]
Mn ₂ CoAl	4.16	7.443	4.05	2.000	0.534	Inv	-0.10	0.29	0.39	НМ	[28,29]
Mn ₂ CuSi	4.06	4.443	4.07	1.000	0.013	Inv	-0.24	0.06	0.30	НМ	[28,29]
Mn ₂ VAl	4.11	2.000	4.19	1.973	-0.963	Reg	0.04	0.40	0.36	Near-HM	[31]
Mn_2FeAs	4.03	3.021	4.04	3.000	0.343	Inv	-0.04	0.21	0.24	НМ	[29]
Mn_2FeSb	4.26	6.350	4.23	3.000	0.310	Inv	-0.03	0.39	0.42	НМ	[28]
Mn ₂ CoSi	4.03	6.280	3.97	3.000	0.730	Inv	-0.25	0.23	0.48	НМ	[28,29]
Mn ₂ CoAs	4.08	5.275	4.06	4.000	0.642	Inv	-0.09	0.12	0.31	НМ	[29]
Mn_2CoGe	4.11	6.470	4.06	3.000	0.570	Inv	-0.11	0.19	0.30	НМ	[28]
Mn ₂ CoSb	4.34	7.570	4.23	4.000	0.590	Inv	-0.01	0.52	0.53	НМ	[28]
Mn_2RhGe	4.32	7.490	4.18	3.000	0.700	Inv	-0.02	0.19	0.21	НМ	[28]
Mn_2 lrGe	4.30	0.000	4.20	3.000	0.700	Inv	-0.01	0.43	0.44	НМ	[28]
Mn ₂ OsSb	4.41	0.010	4.35	3.000	0.830	Inv	0.03	0.35	0.32	Near-HM	[28]
Co ₂ MnSb	4.25	6.000	4.24	5.000	-0.970	Reg	-0.42	0.04	0.46	НМ	[28]
Co ₂ MnSi	3.98	5.000	3.97	4.180	-0.880	Reg	-0.32	0.32	0.64	НМ	[28,31–34]
Co ₂ MnGe	4.06	5.000	4.06	4.610	-0.880	Reg	-0.03	0.37	0.40	НМ	[28,31,34,35]
Co ₂ CrAl	4.03	3.000	4.05	0.841	-0.866	Reg	-0.11	0.43	0.54	НМ	[31,34]
Co ₂ CrSi	3.98	4.000	3.95	9.000	-0.560	Reg	-0.38	0.30	0.68	НМ	[33]
Co ₂ CrGe	4.06	4.000	4.03	0.175	-0.598	Reg	-0.08	0.34	0.42	НМ	[35]
Co ₂ TiAl	4.12	1.000	4.18	2.522	-1.681	Reg	-0.26	0.01	0.27	НМ	[34]
Co ₂ TiSi	4.07	2.000	4.11	1.730	-1.419	Reg	-0.57	0.03	0.60	НМ	[33,34]
Co ₂ VSi	4.01	2.837	4.02	0.803	-0.699	Reg	-0.77	-0.10	0.67	Near-HM	[33,34]
$\rm Co_2Val$	4.06	1.999	4.10	1.817	-1.226	Reg	-0.36	-0.08	0.28	Near-HM	[31]
Fe ₂ CrAl	4.01	0.998	4.09	2.482	-0.394	Reg	-0.21	-0.08	0.13	Near-HM	[31]
Fe_2MnAl	4.01	2.000	4.05	1.930	-0.250	Reg	0.08	0.46	0.38	Near-HM	[31]
Fe ₂ MnSi	3.95	3.000	3.95	3.030	-0.240	Reg	-0.08	0.38	0.46	НМ	[28,31]

For cubic phase $c' = 1/\sqrt{2} \approx 0.707$. Therefore, the deviation of the c'_{opt} from $1/\sqrt{2}$ provides the measure of deviation from cubic structure (the measure of how large is the tetragonal distortion) at a_{opt} . We found that for all the considered compounds in Table 3, K_{mc} is a nearly linear function of *a* that crosses zero at $a = a_c$ due to the cubic symmetry at this value. Therefore, a_{opt} for all the compounds presented in Table 3 coincide either with a_{min} or a_{max} . Thus, the range of lattice constants for which considered full Heusler compounds are simultaneously half-metallic and have PMA is from a_c to a_{opt} . The values of K_{mc} , K_{sh} , and K_v

that correspond to the optimal values of the lattice constants a_{opt} and c'_{opt} are shown in Table 3.

Table 3 is divided into two parts. In the upper part of the table, we show compounds that can be simultaneously half-metals and have PMA ($K_v > 0$) under optimal tetragonal distortions. In the lower part of the table, we show compounds that are not simultaneously half-metals and have PMA for any tetragonal distortion that we considered. While for compounds in the lower part of the table, the largest $K_{\rm mc}$ for *a* in the half-metallicity range is still positive ($K_{\rm mc} > 0$); nevertheless, $K_{\rm mc}$ is smaller than the shape

	a _r [Å]	т, [µ _В]	a _i [Å]	т _і [µ _в]	$E_r - E_i$ [eV]	Ground state			a _r [Å]	т, [µ _В]	a _i [Å]	т _і [µ _В]	$E_r - E_i$ [eV]	Ground state	
Sc ₂ MnAl	4.75	2.560	4.66	2.000	-0.147	Reg	[29]	Cr ₂ CoAl	4.16	5.680	4.05	0.000	0.781	Inv	[29]
Sc ₂ CrSi	4.66	2.740	4.55	2.000	-0.153	Reg	[29]	Cr2MnAs	4.14	0.019	4.13	0.001	-0.206	Reg	[29]
Sc ₂ Vas	4.68	2.071	4.63	2.033	-0.324	Reg	[29]	Cr ₂ FeAs	4.10	0.992	4.07	1.004	0.058	Inv	[29]
Sc_2FeAl	4.62	0.445	4.57	0.936	-0.430	Reg	[29]	Cr ₂ CoAs	4.07	1.569	4.09	2.001	0.398	Inv	[29]
Sc ₂ MnSi	4.53	0.380	4.50	1.000	-0.317	Reg	[29]	Cr₃Al	4.18	2.927					[29]
Sc ₂ CrAs	4.72	2.983	4.59	1.053	-0.579	Reg	[29]	Mn ₂ CrAl	4.05	1.025	4.13	1.000	-0.425	Reg	[29]
Sc ₂ NiAl	4.66	0.000	4.60	0.980	-0.745	Reg	[29]	Mn ₂ CrSi	3.95	0.015	4.04	0.006	-0.356	Reg	[29]
Sc ₂ CoSi	4.55	0.865	4.44	1.000	-0.949	Reg	[29]	Mn ₂ CrAs	4.04	1.004	4.09	1.002	0.117	Inv	[29]
Sc_2Vsi	4.66	2.951	4.62	3.000	-0.276	Reg	[29]	Mn₂ZnSi	4.08	3.666	4.10	2.000	-0.416	Reg	[29]
Sc ₂ CrAl	4.78	3.277	4.72	3.000	-0.215	Reg	[29]	Mn_2VGe	4.07	0.984	4.19	0.903	-0.750	Reg	[31]
Ti ₂ Val	4.48	1.434	4.48	2.000	-0.260	Reg	[29]	Mn ₂ VSi	4.01	0.989	4.06	0.947	-1.127	Reg	[31]
Ti ₂ Vsi	4.36	0.000	4.37	1.000	-0.264	Reg	[29]	Mn_2VSn	4.26	0.998	4.44	4.587	-0.503	Reg	[31]
Ti ₂ MnAl	4.36	0.003	4.42	0.000	-0.196	Reg	[29]	Co ₂ MnAl	4.03	4.010	4.07	5.250	-0.830	Reg	[31]
Ti ₂ Vas	4.39	0.000	4.40	0.001	-0.197	Reg	[29]	Co₂MnGa	4.05	4.080	4.09	5.380	-0.690	Reg	[31]
Ti ₂ FeAl	4.38	0.985	4.34	1.000	-0.135	Reg	[29]	Co ₂ MnSn	4.23	5.010	4.27	5.630	-0.970	Reg	[31]
Ti ₂ CoAl	4.38	2.000	4.34	2.000	-0.092	Reg	[29]	Co_2FeAl	4.03	4.980	4.02	4.390	-0.690	Reg	[34]
Ti ₂ FeSi	4.31	1.884	4.24	2.000	-0.242	Reg	[29]	Co₂FeSi	3.98	5.460	3.95	3.960	-0.380	Reg	[32,33]
Ti ₂ NiAl	4.40	1.391	4.38	3.000	-0.047	Reg	[29]	Co₂FeGa	4.04	5.000	4.05	4.800	-0.630	Reg	[34]
Ti₂CoSi	4.30	2.327	4.26	3.000	-0.272	Reg	[29]	Co ₂ FeGe	4.06	5.630	4.03	4.230	-0.440	Reg	[34]
V ₃ Al	4.27	0.000					[29]	Co₂CrGa	4.05	3.008	4.07	0.658	-0.673	Reg	[35,36]
V_2CrAl	4.23	0.986	4.24	0.938	-0.050	Reg	[29]	Fe ₂ VAl	4.03	0.000	4.14	3.572	-1.185	Reg	[31]
V ₂ CoAl	4.22	3.335	4.18	2.000	0.584	Inv	[29]	Ru ₂ MnSi	4.18	3.002	4.17	1.437	-1.874	Reg	[31]
V_2 FeSi	4.15	1.040	4.10	1.997	0.331	Inv	[29]	Ru₂MnGe	4.25	3.030	4.23	1.340	-1.740	Reg	[31]
V_2MnAs	4.23	0.319	4.17	1.986	0.047	Inv	[29]	Ru₂MnSn	4.41	3.070	4.44	4.340	-1.390	Reg	[31]
V ₂ CoSi	4.13	2.120	4.09	0.999	0.330	Inv	[29]	Rh_2MnAl	4.27	4.039	4.31	4.018	-1.768	Reg	[31]
V_2FeAs	4.20	1.556	4.14	1.000	-0.155	Reg	[29]	Rh₂MnGa	4.28	4.090	4.33	4.020	-1.540	Reg	[31]
Cr_2FeAl	4.17	0.947	4.07	1.005	0.619	Inv	[29]	Ru ₂ MnSb	4.41	4.010	4.45	4.480	-1.640	Reg	[31]
Cr₃As	4.13	0.990					[29]								

anisotropy constant, $K_{\rm sh}$, so that $K_{\rm v} = K_{\rm mc} - K_{\rm sh}$ is negative. Unfortunately, one of the most well-known half-metallic Heusler compounds, Co2MnSi, belongs to the lower list of compounds that cannot be simultaneously half-metallic and have PMA.

Compounds in the upper and lower parts of Table 3 are listed in order of the magnitude of the width of the half-metallicity range, $w_{\rm hm}$. Six compounds from the top of Table 3, namely, Mn2CoSi, Mn2CoAl, Co2CrSi, Mn2CuSi, Mn2CoGe, and Co₂CrGe, have a wide half-metallicity range, $w_{\rm hm} \ge 0.24$ Å, a relatively large bandgap, $E_{\rm gap} \ge 0.30 \, {\rm eV}$, a large Curie temperature, $T_{\rm C} \ge 490$ K, and a relatively large PMA under the optimal tetragonal distortion, $K_v \ge 0.18 \text{ MJ m}^{-3}$. These six compounds therefore form our list of the most promising full-Heusler compounds for MTJ devices.

Note that Mn₂FeSb is listed in Table 3 twice. The reason is that Mn₂FeSb has two ranges of a where it is half-metallic. One half-metallicity range has $a_{\min} = 4.17$ Å and $a_{\max} = 4.39$ Å (in this range the magnetic moment is $3.0 \mu_{\rm B}$ per formula unit). The second half-metallicity range corresponds to $a_{\min} = 4.03 \text{ Å}$ and $a_{\text{max}} = 4.05$ Å (in this range the magnetic moment is $1.0 \,\mu_{\text{B}}$ per formula unit). While the width of the second half-metallicity range is only $w_{\rm hm} = 0.02$ Å, we nevertheless include the second entry for Mn₂FeSb in Table 3 for two reasons. The first reason is that due to a stronger tetragonal distortion the optimal K_v increases from $K_v = 0.04 \text{ MJ m}^{-3}$ for the first range to $K_v = 1.15 \text{ MJ m}^{-3}$ for the second range (this is the largest value of K_v presented in Table 3). The second reason is that $a_{\rm opt} = 4.03$ Å ideally matches the lattice constant of a CoAl underlayer material (a = 4.04 Å). It has been shown recently that using a novel chemical templating technique, the use of a chemical template underlayer formed from an atomically ordered CoAl layer allows for near-bulk-like magnetic properties in tetragonally distorted Heusler films grown on top of it, even for film deposition at room temperature, and that films only 1 or 2 unit cells thick display excellent PMA properties.[14,30]

ANCED

regular and inverse structures, $E_r - E_i$.

www.advancedsciencenews.com



Table 3. Lattice constant, a_c , total magnetic moment, m_c , the bandgap, E_{gap} , and the Curie temperature, T_C , calculated for the ground state cubic structure of half-metallic full-Heuslers, the minimum, a_{min} , and the maximum, a_{max} , values of the in-plane lattice constant for which the tetragonally distorted compound is staying half-metallic, the width of the half-metallic range, $w_{hm} = a_{max} - a_{min}$, the optimal in-plane lattice constant, a_{opt} , and corresponding dimensionless out-of-plane lattice constant, c'_{opt} , that provide the largest (optimal) MCA energy for a in the half-metallicity range ($a_{min} \le a_{opt} \le a_{max}$), the MCA energy, K_{mcr} , shape anisotropy energy, K_{sh} , and the volume magnetic anisotropy, $K_v = K_{mc} - K_{sh}$, calculated for the optimal lattice constant, a_{opt} (PMA corresponds to positive K_v). Top part of the table contains compounds with PMA ($K_v < 0$). In both parts of the table, compounds are ordered by the width of the half-metallic range, w_{hm} .

	Ground state	a _c [Å]	т _с [µ _В]	Egap [eV]	<i>T</i> _C [K]	a _{min} [Å]	a _{max} [Å]	w _{hm} [Å]	a _{opt} [Å]	$c'_{\rm opt}$	$K_{\rm mc} [{\rm meV} {\rm f.u.}^{-1}]$	K _{sh} [meV f.u. ⁻¹]	$K_{\rm v} [{\rm MJ} {\rm m}^{-3}]$
Mn ₂ CoSi	Inv	3.97	3.00	0.48	648	3.83	4.27	0.44	3.83	0.767	0.22	0.07	0.54
Mn ₂ CoAl	Inv	4.05	2.00	0.39	872	3.91	4.25	0.34	3.91	0.761	0.09	0.03	0.23
Co ₂ CrSi	Reg	3.98	4.00	0.68	567	3.88	4.22	0.34	3.88	0.753	0.17	0.12	0.18
Mn ₂ CuSi	Inv	4.07	1.00	0.30	690	3.97	4.29	0.32	4.29	0.619	0.14	0.01	0.43
Mn_2CoGe	Inv	4.06	3.00	0.30	623	3.96	4.26	0.30	3.96	0.747	0.15	0.07	0.28
Co ₂ CrAl	Reg	4.03	3.00	0.54	388	3.89	4.19	0.30	3.89	0.771	0.35	0.07	0.99
Co ₂ CrGe	Reg	4.06	4.00	0.42	492	3.94	4.18	0.24	3.94	0.759	0.21	0.12	0.32
Mn_2FeSb	Inv	4.23	3.00	0.42	411	4.17	4.39	0.22	4.17	0.731	0.07	0.06	0.04
Mn_2FeSb	Inv	4.23	3.00	0.42	411	4.03	4.05	0.02	4.03	0.858	0.41	0.01	1.15
Mn₃Al		4.09	0.00	0.51	333	3.97	4.15	0.18	3.97	0.763	0.25	0.00	0.84
Mn ₂ CoSb	Inv	4.23	4.00	0.53	656	4.11	4.27	0.16	4.11	0.756	0.11	0.10	0.03
$V_2 MnAl$	Inv	4.20	2.00	0.08	360	4.16	4.32	0.16	4.32	0.670	0.11	0.02	0.26
Cr ₂ ZnSi	Inv	4.13	0.00	0.43	233	3.97	4.03	0.06	3.97	0.816	0.14	0.00	0.45
Co ₂ TiSi	Reg	4.07	2.00	0.60	94	4.05	4.09	0.04	4.05	0.713	0.06	0.03	0.11
Co_2MnSi	Reg	3.98	5.00	0.64	970	3.86	4.26	0.40	3.86	0.755	0.13	0.19	-0.26
Co_2MnSb	Reg	4.25	6.00	0.46	788	4.23	4.55	0.32	4.23	0.717	0.06	0.22	-0.49
Mn_2CoAs	Inv	4.06	4.00	0.31	692	3.92	4.14	0.22	3.92	0.766	0.06	0.12	-0.20
Co_2MnGe	Reg	4.06	5.00	0.40	866	3.90	4.10	0.20	3.90	0.773	0.13	0.18	-0.19
Fe ₂ MnSi	Reg	3.95	3.00	0.46	168	3.85	4.05	0.20	4.05	0.666	0.01	0.07	-0.20
Mn_2FeAs	Inv	4.04	3.00	0.24	563	3.98	4.14	0.16	3.98	0.735	0.07	0.07	0.00
Mn_2 lrGe	Inv	4.20	3.00	0.44	345	4.18	4.24	0.06	4.18	0.720	0.05	0.06	-0.02
Mn_2RhGe	Inv	4.18	3.00	0.21	368	4.16	4.22	0.06	4.16	0.720	0.01	0.06	-0.15
Co ₂ TiAl	Reg	4.12	1.00	0.27	54	4.12	4.16	0.04	4.12	0.707	0.00	0.01	-0.02

3. Half-Metallic and PMA Properties of Half-Heusler Compounds

3.1. Crystal Structure

The crystal structure of the cubic half-Heusler XYZ compounds can be obtained from the crystal structure of the full-Heusler X_2YZ compounds shown in Figure 1c as follows: the *fcc* sublattice of Z-atoms stays intact, but one of the remaining three *fcc* sublattices of atoms represented by either blue, red, or orange spheres in the figure is removed and the remaining two *fcc* sublattices are occupied by X and Y atoms. There are three possible resulting structures: we will use notation indicators 1, 2, and 3 for these three possible crystal structures of half-Heusler compounds as follows: 1) 1 denotes the crystal structure when the X atoms are located in one *xy*-plane and Y and Z atoms are located in a second *xy*-plane; 2) 2 denotes the crystal structure when Y atoms are located in one *xy*-plane and X and Z atoms are located in a second *xy*-plane; and 3) 3 denotes the crystal structure when Z atoms are located in one *xy*-plane and X and Y atoms are located in a second *xy*-plane.

Analogous to the case of full-Heuslers a tetragonal half-Heusler structure can be obtained from the cubic structure by stretching (or compressing) the parent cubic structure along the *z*-axis.

We performed DFT calculations for all three possible crystal structures with various initial magnetic configurations and various cubic lattice constants for 147 cubic half-Heusler compounds that were identified as half-metals in the literature. For DFT calculations we used the computational approach described in Section 2.2. For selected compounds (see Table 6 and 7), we also performed calculations for tetragonally distorted structures.

3.2. Results and Discussion

The results of calculations for half-Heuslers compounds are summarized in **Table 4–7**. We found that out of 147 considered

ADVANCED SCIENCE NEWS _____



Table 4. Ground state structure indicator (see definition of the three possible structures of half-Heuslers in the text) of half-Heuslers that we identified as half-metals or near-half-metals, calculated cubic lattice constant, a_c , and total magnetic moment, m_c , in the ground state, the indicator of the crystal structure with the second lowest total energy, the difference, $E_1 - E_2$, between the total energy of the ground state crystal structure, E_1 , and the total energy of the crystal structure with the second lowest total energy, E_2 , the bottom edge of the bands gap, E_{min} (where we set $E_F = 0$), top edge of the bandgap, $E_{max} - E_{min}$, calculated for the ground state structure, the spin channel which has the bandgap. Note that CoTiSb has bandgaps in both spin channels, so it is a semiconductor, not a half-metal.

	Ground state	a _c [Å]	m _c [μ _B]	Second lowest state	$E_1 - E_2$ [eV]	E _{min} [eV]	E _{max} [eV]	E_{gap} [eV]	Gap spin channel		
NiMnSb	1	4.18	4.000	2	-0.955	-0.163	0.327	0.490	2	НМ	[37-40]
NiVSb	1	4.17	2.000	3	-0.945	-0.368	0.067	0.435	2	НМ	[39,40]
NaKSb	3	5.65	1.000	1	-0.130	-0.310	1.342	1.652	1	НМ	[41]
PtMnSb	1	4.41	4.017	2	-0.805	0.108	0.769	0.661	2	Near-HM	[37,39]
FeMnSb	1	4.13	2.000	2	-0.565	-0.206	0.778	0.984	2	НМ	[39]
MnVSb	1	4.20	1.000	2	-0.931	-0.013	0.836	0.849	1	НМ	[40]
CrScSb	1	4.56	4.000	3	-0.215	-0.265	0.710	0.975	1	НМ	[40]
CoMnSb	1	4.11	3.000	2	-0.948	-0.096	0.827	0.923	2	НМ	[38-40,42
CoVSb	1	4.12	1.000	2	-1.373	-0.791	0.134	0.925	2	НМ	[39,40,42
CoCrSb	1	4.11	2.000	2	-0.996	-0.221	0.592	0.813	2	НМ	[39,42]
CoTiSb	1	4.17	0.000	2	-2.286	-0.004	1.044	1.048	1,2	semi	[43]
RhVSb	1	4.30	1.000	2	-0.842	-0.499	0.360	0.859	2	НМ	[40]
KCrS	3	4.79	5.000	2	-0.353	-2.939	-0.012	2.927	2	Near-HM	[44]
RbCrS	3	4.97	5.000	2	-0.208	-2.416	0.021	2.437	2	НМ	[44]
CsCrS	3	5.19	5.000	2	-0.022	-1.809	0.223	2.032	2	НМ	[44]
CsVS	2	4.91	4.000	3	-0.093	-2.597	0.205	2.802	2	НМ	[45]
NaKP	3	5.22	1.000	1	-0.298	-0.542	1.428	1.970	1	НМ	[41]
NaCsP	3	5.54	1.000	1	-0.221	-0.342	1.142	1.484	1	НМ	[46]
CsRbP	3	5.88	1.000	2	-0.603	-0.366	1.222	1.588	1	НМ	[47]
CoVP	1	3.80	0.999	3	-0.376	-1.416	-0.079	1.337	2	Near-HM	[40]
LiCaB	3	4.40	2.000	1	-0.770	-0.010	1.153	1.163	1	НМ	[48]
LiSrB	3	4.65	2.000	1	-0.761	-0.066	1.091	1.157	1	НМ	[48]
NaSrB	3	4.85	2.000	1	-1.225	-0.041	0.995	1.036	1	НМ	[48]
NaBaB	3	5.07	1.968	1	-1.182	0.037	0.876	0.839	1	Near-HM	[48]
RbBaB	3	5.45	1.999	2	-1.046	0.016	0.633	0.617	1	Near-HM	[48]
NaCaB	3	4.63	2.000	2	-1.294	-0.062	1.041	1.103	1	НМ	[48]
КсаВ	3	4.98	2.000	2	-1.018	-0.172	0.700	0.872	1	НМ	[48]
KSrB	3	5.16	2.000	2	-1.148	-0.156	0.668	0.824	1	НМ	[48]
KbaB	3	5.33	2.000	2	-1.122	-0.054	0.627	0.681	1	НМ	[48]
RbCaB	3	5.14	2.000	2	-0.931	-0.084	0.696	0.780	1	НМ	[48]
RbSrB	3	5.30	2.000	2	-1.075	-0.096	0.649	0.745	1	НМ	[48]
MgCaB	3	4.43	1.000	1	-1.686	-0.243	0.522	0.765	1	НМ	[49]
CsVSe	2	5.06	4.000	3	-0.232	-2.421	0.233	2.654	2	НМ	[45]
CsCrSe	2	5.14	5.000	3	-0.097	-1.801	0.750	2.551	2	НМ	[44]
RbCrSe	3	5.14	5.000	2	-0.071	-2.290	-0.098	2.192	2	Near-HM	[44]
FeCrSe	1	3.96	2.000	2	-0.397	-0.656	-0.012	0.644	2	Near-HM	[50]
LiCaGe	3	4.84	1.000	1	-0.127	-0.116	1.089	1.205	1	НМ	[51,52]
LiSrGe	3	5.07	1.000	1	-0.107	-0.327	1.049	1.376	1	НМ	[52]
LiBaGe	3	5.28	1.000	1	-0.123	-0.046	1.093	1.139	1	НМ	[52]
LiVGe	3	4.19	2.000	1	-0.764	-0.743	0.036	0.779	2	НМ	[45]
NaCaGe	3	5.07	1.000	1	-0.769	-0.256	1.014	1.270	1	НМ	[51]
KMgGe	3	5.14	1.000	2	-0.243	-0.079	0.202	0.281	1	НМ	[51]
KCaGe	3	5.38	1.000	2	-0.644	-0.348	0.769	1.117	1	НМ	[51,53]

ADVANCED SCIENCE NEWS

www.advancedsciencenews.com

Table 4. Continued.



	Ground state	a _c [Å]	т _с [µ _В]	Second lowest state	$E_1 - E_2$ [eV]	E _{min} [eV]	E _{max} [eV]	Egap [eV]	Gap spin channel		
RbTaGe	3	4.95	2.000	2	-0.052	-0.332	0.297	0.629	2	НМ	[45]
RbSrGe	3	5.68	1.000	2	-0.714	-0.198	0.410	0.608	1	НМ	[54]
CsNbGe	2	4.94	2.000	3	-0.558	-0.235	0.099	0.334	2	НМ	[45]
CsSrGe	3	5.81	1.000	2	-0.575	-0.041	0.350	0.391	1	НМ	[55]
CsBaGe	3	5.94	1.000	2	-0.587	0.008	0.517	0.509	1	Near-HM	[56]
RhFeGe	1	4.09	3.000	3	-0.248	-0.396	0.014	0.410	2	НМ	[40,57]
RhCrGe	1	4.07	1.000	3	-0.158	-0.401	0.277	0.678	2	НМ	[40]
NiCrGe	1	3.93	2.000	3	-0.296	-0.130	0.544	0.674	2	НМ	[40]
NiVGe	1	3.95	0.996	3	-0.742	-0.801	-0.071	0.730	2	Near-HM	[40]
CoFeGe	1	3.89	3.000	2	-0.496	-0.263	0.122	0.385	2	НМ	[40]
CoCrGe	1	3.88	1.000	3	-0.675	-0.705	0.291	0.996	2	НМ	[40]
CoMnGe	1	3.89	2.000	3	-0.486	-0.156	0.855	1.011	2	НМ	[40]
YMnGe	3	4.58	4.000	2	-0.499	-0.043	0.271	0.314	1	НМ	[58]
NaVTe	3	4.84	4.000	2	-0.140	-3.078	0.159	3.237	2	НМ	[45]
KCrTe	2	5.06	5.000	3	-0.110	-2.317	0.520	2.837	2	НМ	[44]
RbVTe	2	5.15	4.000	3	-0.200	-2.315	0.498	2.813	2	НМ	[45]
RbNbTe	2	5.25	4.000	3	-0.008	-2.720	0.201	2.921	2	НМ	[45]
RbCrTe	2	5.20	5.000	3	-0.201	-2.012	0.589	2.601	2	НМ	[44]
CsCrTe	2	5.37	5.000	3	-0.329	-1.627	0.636	2.263	2	НМ	[44]
CsNbTe	2	5.36	3.997	3	-0.300	-2.491	-0.085	2.406	2	Near-HM	[45]
NaCaSn	3	5.34	1.000	1	-0.503	-0.124	1.013	1.137	1	НМ	[51]
KTaSn	3	5.08	2.000	2	-0.120	-0.370	0.314	0.684	2	НМ	[45]
KCaSn	3	5.66	1.000	2	-0.482	-0.310	0.884	1.194	1	НМ	[51,53]
RbNbSn	2	5.10	2.000	3	-0.266	-0.269	0.461	0.730	2	НМ	[45]
CsNbSn	2	5.17	2.000	3	-0.693	-0.132	0.119	0.251	2	НМ	[45]
CsSrSn	3	6.09	1.000	2	-0.440	-0.070	0.576	0.646	1	НМ	[55]
RhFeSn	1	4.28	3.000	2	-0.428	-0.226	0.192	0.418	2	НМ	[40]
NiVSn	1	4.15	1.000	2	-1.366	-0.485	0.018	0.503	2	НМ	[40]
RbTaSn	2	5.08	1.997	3	-0.175	0.010	0.028	0.018	1	Near-HM	[45]
YMnSn	2	4.71	3.986	3	-0.260	0.109	0.380	0.271	1	Near-HM	[58]
NaVSi	3	4.42	2.000	2	-0.489	-0.418	0.391	0.809	2	НМ	[45]
KVSi	2	4.64	2.000	3	-0.060	-0.084	0.924	1.008	2	НМ	[45]
KcaSi	3	5.34	1.000	2	-0.644	-0.362	0.882	1.244	1	НМ	[51]
RbSrSi	3	5.63	1.000	2	-0.715	-0.203	0.785	0.988	1	НМ	[54]
RbNbSi	2	4.79	2.000	3	-0.134	-0.494	0.308	0.802	2	НМ	[45]
RbTaSi	3	4.87	2.000	2	-0.036	-0.396	0.207	0.603	2	НМ	[45]
CsNbSi	2	4.88	2.000	3	-0.589	-0.175	0.017	0.192	2	НМ	[45]
NiCrSi	1	3.85	2.000	3	-0.173	-0.439	0.438	0.877	2	НМ	[40]
NiMnSi	1	3.86	3.000	3	-0.271	-0.309	0.565	0.874	2	НМ	[40,53,57]
CoMnSi	1	3.80	2.000	3	-0.311	-0.301	0.503	0.804	2	НМ	[40]
CoCrSi	1	3.80	1.000	3	-0.812	-0.847	-0.006	0.841	2	Near-HM	[40]
CoTiSi	1	3.94	1.000	3	-1.084	0.008	1.194	1.186	-	Near-HM	[57]
YMnSi	3	4,52	3,981	2	-0,511	0.086	0.315	0.229	1	Near-HM	[58]
NiCrAs	3	3.97	3,000	-	-0.017	-0.127	0.466	0.593	2	НМ	[40]
NiMnAs	1	3.99	4,000	3	-0.479	-0.284	0.429	0.713	2	нм	[40]
NaKAs	3	5.35	1.000	1	-0.253	-0.464	1,141	1.605	-	нм	[41]
NaCsAs	3	5.68	1,000	1	-0,177	-0,334	0.905	1.239	1	нм	[46]
	-										1.01

ADVANCED SCIENCE NEWS

www.advancedsciencenews.com

Table 4. Continued.



	Ground state	a _c [Å]	m _c [μ _B]	Second lowest state	$E_1 - E_2$ [eV]	E _{min} [eV]	E _{max} [eV]	E_{gap} [eV]	Gap spin channel		
FeMnAs	1	3.90	2.000	2	-0.423	-0.464	0.621	1.085	2	НМ	[40]
RhVAs	1	4.12	1.000	3	-0.406	-0.699	0.218	0.917	2	НМ	[40]
RuMnAs	1	4.08	2.000	3	-0.102	-0.016	0.747	0.763	2	НМ	[40,57]
MnMnAs	1	3.97	1.000	3	-0.387	-0.594	0.505	1.099	2	НМ	[40]
FeCrAs	1	3.88	1.000	2	-0.606	-0.748	0.250	0.998	2	НМ	[40]
RuCrAs	1	4.07	1.000	2	-0.148	-0.348	0.230	0.578	2	НМ	[40]
CoVAs	1	3.92	1.000	3	-0.952	-1.139	0.104	1.243	2	НМ	[40]
CoCrAs	1	3.91	2.000	3	-0.392	-0.597	0.524	1.121	2	НМ	[40]
CoMnAs	1	3.91	3.000	3	-0.532	-0.405	0.805	1.210	2	НМ	[40]
MnVAs	1	3.97	1.000	3	-0.602	-0.065	0.761	0.826	1	НМ	[40]
CsRbAs	3	6.03	1.000	2	-0.537	-0.339	1.019	1.358	1	НМ	[47]
CsBaC	3	5.22	1.000	2	-1.163	-0.333	0.419	0.752	1	НМ	[56]
CsSrC	3	5.10	1.000	2	-1.117	-0.251	0.197	0.448	1	НМ	[56]
RbSrC	3	4.97	1.000	2	-1.177	-0.377	0.208	0.585	1	НМ	[54]
LiCaC	3	4.15	1.000	1	-1.067	-0.666	1.500	2.166	1	НМ	[48]
NaCaC	3	4.36	1.000	2	-1.534	-0.632	1.127	1.759	1	НМ	[48]
KcaC	3	4.68	1.000	2	-1.087	-0.447	0.687	1.134	1	НМ	[49]
NaKN	3	4.53	1.000	1	-0.700	-1.020	0.808	1.828	1	НМ	[41]
CsRbN	3	5.20	1.000	2	-1.119	-0.814	0.649	1.463	1	НМ	[47]
NiFeGa	1	3.93	3.000	2	-0.597	-0.434	0.124	0.558	2	НМ	[40]
NiTiIn	1	4.25	0.993	2	-1.617	0.026	0.506	0.480	1	Near-HM	[40]

Table 5.	Ground state structure indicator of half-Heuslers that we identified as not half-metals or near-half-metals, calculated cubic lattice constant, a	1 _c ,
and tota	magnetic moment, m_{c} , in the ground state, the indicator of the crystal structure with the second lowest total energy, the difference, $E_1 - E_2$	2,
between	the total energy of the ground state crystal structure, E_1 , and the total energy of the crystal structure with the second lowest total energy, E_1	2.

	Ground state	a _c [Å]	m _c [μ _B]	Second lowest state	$E_1 - E_2$ [eV]			Ground state	a _c [Å]	m _c [μ _B]	Second lowest state	$E_1 - E_2$ [eV]	
NiCrSb	1	4.17	3.027	3	-0.796	[39]	LiBaB	3	4.88	1.763	1	-0.757	[48]
PdMnSb	1	4.40	4.058	2	-0.545	[39]	KcrSe	3	4.96	4.999	2	-0.184	[44]
IrMnSb	1	4.32	3.041	2	-1.017	[39]	LiCrSe	3	4.35	4.867	2	-0.449	[53]
LiCrS	3	4.09	4.447	1	-1.022	[59]	RbTaTe	2	5.22	3.859	3	-0.054	[45]
LiMnP	3	4.03	4.097	2	-0.670	[60]	LiMnSi	3	4.08	3.318	2	-0.408	[60]
FeMnP	3	3.74	3.280	1	-0.059	[40]	RhCrSi	3	4.00	3.067	1	-0.044	[40]
CoMnP	3	3.73	2.417	1	-0.208	[57]	LiVSi	3	4.13	1.979	1	-0.804	[45]
RhMnP	3	3.89	2.550	1	-0.620	[40]	NiVSi	1	3.88	0.969	3	-0.659	[40]
RhCrP	3	3.93	2.725	1	-0.795	[40]	FeMnSi	1	3.77	0.979	3	-0.320	[40]
RhVP	3	3.96	1.528	1	-0.212	[40]	RhCrAs	3	4.08	2.918	1	-0.182	[40]
CoCrP	3	3.78	2.642	1	-0.214	[40]	CrMnAs	2	3.87	0.002	3	-0.401	[40]
NiCrP	3	3.81	2.535	1	-0.653	[40]	CrScAs	3	4.36	3.714	1	-0.620	[40]
NiMnP	3	3.72	1.296	1	-0.461	[57]	CrTiAs	3	4.16	2.131	1	-0.636	[40]
CrMnP	2	3.75	0.000	3	-0.290	[40]	FeFeAs	1	3.88	2.994	3	-0.531	[40]
CrScP	3	4.32	3.669	1	-0.788	[40]	RuMnIn	1	4.49	4.067	2	-0.026	[40]
MnMnP	3	3.79	4.422	1	-0.166	[40]	LiMnN	3	3.49	4.001	2	-2.004	[60]
RuCrP	3	3.91	2.522	1	-0.191	[40]	PtMnBi	1	4.53	4.119	2	-0.679	[61]
RuMnP	3	3.89	2.838	1	-0.598	[40]	CoMnAl	1	3.86	1.016	2	-0.758	[40]



Table 6. Ground state structure indicator, lattice constant, a_c , total magnetic moment, m_c , the bandgap, E_{gap} , and the Curie temperature, T_c , calculated for the ground state cubic structure of half-metallic half-Heuslers, the minimum, a_{min} , and the maximum, a_{max} , values of the in-plane lattice constant for which the tetragonally distorted compound stays half-metallic, the width of the half-metallic range, $w_{hm} = a_{max} - a_{min}$, the optimal in-plane lattice constant, a_{opt} , and corresponding dimensionless out-of-plane lattice constant, c'_{opt} , that provide the largest (optimal) MCA energy for a in the half-metallicity range ($a_{min} \leq a_{opt} \leq a_{max}$), the MCA energy, K_{mc} , shape anisotropy energy, K_{sh} , and the volume magnetic anisotropy, $K_v = K_{mc} - K_{sh}$, calculated for the optimal lattice constant, a_{opt} . This table includes compounds that have PMA ($K_v > 0$). Compounds are ordered by the width of the half-metallic range, w_{hm} . The bold font for some values of a_{min} and a_{max} indicates that the true value of a_{min} is smaller and the true value of a_{max} is larger than the values shown in table (see text for details).

	Ground state	a _c [Å]	m _c [μ _B]	Egap [eV]	<i>T</i> _C [K]	a _{min} [Å]	a _{max} [Å]	w _{hm} [Å]	a _{opt} [Å]	$c'_{\rm opt}$	K _{mc} [meV f.u. ⁻¹]	$K_{\rm sh} \ [{\rm meV} \ {\rm f.u.}^{-1}]$	$K_{\rm v} [{\rm MJ} {\rm m}^{-3}]$
KCrTe	2	5.06	5.000	2.837	155	4.48	5.72	1.24	4.48	0.889	0.308	0.105	0.406
NaCsP	3	5.54	1.000	1.484	313	4.94	6.14	1.20	4.94	0.877	0.249	0.003	0.372
NaCsAs	3	5.68	1.000	1.239	291	5.08	6.28	1.20	5.10	0.887	0.943	0.003	1.281
CsRbAs	3	6.03	1.000	1.358	345	5.43	6.63	1.20	5.57	0.817	0.338	0.002	0.381
CsRbN	3	5.20	1.000	1.463	118	4.60	5.80	1.20	5.80	0.630	0.157	0.003	0.201
NaKP	3	5.22	1.000	1.970	341	4.62	5.82	1.20	5.82	0.590	0.006	0.003	0.004
CsRbP	3	5.88	1.000	1.588	289	5.28	6.44	1.16	6.38	0.624	0.062	0.002	0.059
KCaGe	3	5.38	1.000	1.117	260	4.96	6.04	1.08	6.02	0.615	0.318	0.003	0.377
RbCrTe	2	5.20	5.000	2.601	113	4.64	5.72	1.08	4.74	0.844	0.166	0.094	0.129
CsBaC	3	5.22	1.000	0.752	417	4.74	5.78	1.04	4.98	0.754	0.259	0.004	0.439
LiSrGe	3	5.07	1.000	1.376	253	4.75	5.73	0.98	5.71	0.585	0.172	0.003	0.248
RbSrGe	3	5.68	1.000	0.608	265	5.30	6.28	0.98	6.28	0.613	0.028	0.002	0.028
KCaSn	3	5.66	1.000	1.194	217	5.28	6.26	0.98	6.24	0.608	1.279	0.002	1.386
KCaSi	3	5.34	1.000	1.244	283	4.90	5.82	0.92	5.18	0.750	0.008	0.003	0.007
NaVSi	3	4.42	2.000	0.809	676	4.08	4.98	0.90	4.94	0.596	0.087	0.019	0.152
RbSrSi	3	5.63	1.000	0.988	287	5.19	6.09	0.90	6.09	0.629	0.058	0.002	0.063
RbSrB	3	5.30	2.000	0.745	450	5.08	5.94	0.86	5.08	0.743	0.046	0.014	0.053
NaCaGe	3	5.07	1.000	1.270	267	4.79	5.61	0.82	5.59	0.608	0.235	0.003	0.349
KSrB	3	5.16	2.000	0.824	509	4.94	5.76	0.82	4.94	0.746	0.021	0.015	0.011
KTaSn	3	5.08	2.000	0.684	236	4.80	5.60	0.80	5.56	0.599	8.940	0.013	13.885
RbTaGe	3	4.95	2.000	0.629	510	4.77	5.55	0.78	5.37	0.624	6.474	0.014	10.715
CsSrSn	3	6.09	1.000	0.646	240	5.95	6.73	0.78	6.71	0.609	0.697	0.002	0.606
CsVS	2	4.91	4.000	2.802	19	4.77	5.55	0.78	5.45	0.617	0.140	0.054	0.138
RbSrC	3	4.97	1.000	0.585	210	4.37	5.15	0.78	4.79	0.740	0.020	0.004	0.032
CsSrC	3	5.10	1.000	0.448	420	4.52	5.28	0.76	4.84	0.757	0.156	0.004	0.284
RbTaSi	3	4.87	2.000	0.603	450	4.75	5.51	0.76	5.27	0.628	6.100	0.015	10.608
KBaB	3	5.33	2.000	0.681	301	5.25	5.97	0.72	5.25	0.720	0.070	0.013	0.088
CoVSb	1	4.12	1.000	0.925	204	3.74	4.46	0.72	4.28	0.661	0.026	0.007	0.060
RbNbSn	2	5.10	2.000	0.730	658	4.62	5.34	0.72	5.34	0.666	0.049	0.013	0.056
RbNbSi	2	4.79	2.000	0.802	707	4.57	5.25	0.68	4.57	0.764	0.146	0.018	0.280
KMgGe	3	5.14	1.000	0.281	280	4.92	5.60	0.68	5.60	0.627	0.060	0.003	0.082
MnMnAs	1	3.97	1.000	1.099	1341	3.69	4.35	0.66	3.81	0.765	0.009	0.008	0.004
RbCaB	3	5.14	2.000	0.780	488	5.00	5.64	0.64	5.00	0.733	0.036	0.015	0.037
CsSrGe	3	5.81	1.000	0.391	280	5.71	6.31	0.60	5.73	0.727	0.037	0.002	0.041
CoCrAs	1	3.91	2.000	1.121	794	3.57	4.15	0.58	3.57	0.869	0.095	0.034	0.247
LiBaGe	3	5.28	1.000	1.139	195	5.20	5.78	0.58	5.20	0.730	0.061	0.003	0.090
CoCrSb	1	4.11	2.000	0.813	678	3.63	4.19	0.56	4.19	0.682	0.064	0.027	0.119
NaCaSn	3	5.34	1.000	1.137	216	5.16	5.72	0.56	5.72	0.625	0.825	0.003	1.126
CoCrGe	1	3.88	1.000	0.996	337	3.64	4.18	0.54	3.64	0.816	0.056	0.009	0.193
RhVAs	1	4.12	1.000	0.917	423	3.98	4.52	0.54	4.02	0.756	0.017	0.007	0.033
CsNbSn	2	5.17	2.000	0.251	620	5.01	5.53	0.52	5.53	0.658	0.115	0.012	0.148

ADVANCED SCIENCE NEWS

www.advancedsciencenews.com

Table 6. Continued.



	Ground state	a _c [Å]	т _с [µ _В]	E_{gap} [eV]	<i>T</i> _C [K]	a _{min} [Å]	a _{max} [Å]	w _{hm} [Å]	a _{opt} [Å]	$c'_{\rm opt}$	$K_{\rm mc}$ [meV f.u. ⁻¹]	$K_{\rm sh} \ [{\rm meV f.u.}^{-1}]$	$K_{\rm v} [{\rm MJ} {\rm m}^{-3}]$
CsNbSi	2	4.88	2.000	0.192	546	4.88	5.40	0.52	5.38	0.628	0.079	0.014	0.107
CsNbGe	2	4.94	2.000	0.334	571	4.86	5.36	0.50	5.36	0.644	0.084	0.014	0.114
NiMnSi	1	3.86	3.000	0.874	1420	3.50	4.00	0.50	3.50	0.899	0.089	0.079	0.043
RhCrGe	1	4.07	1.000	0.678	82	3.81	4.31	0.50	3.81	0.837	0.148	0.007	0.487
LiCaGe	3	4.84	1.000	1.205	239	4.70	5.18	0.48	5.18	0.627	0.166	0.004	0.298
LiSrB	3	4.65	2.000	1.157	538	4.59	5.05	0.46	4.59	0.720	0.025	0.019	0.013
CoMnGe	1	3.89	2.000	1.011	320	3.57	4.01	0.44	4.01	0.666	0.076	0.031	0.166
CoMnSi	1	3.80	2.000	0.804	523	3.60	4.04	0.44	3.70	0.751	0.042	0.035	0.028
FeCrAs	1	3.88	1.000	0.998	532	3.78	4.20	0.42	3.78	0.745	0.046	0.008	0.150
RuCrAs	1	4.07	1.000	0.578	267	3.95	4.37	0.42	3.95	0.759	0.071	0.007	0.219
KVSi	2	4.64	2.000	1.008	310	4.42	4.84	0.42	4.42	0.758	0.038	0.021	0.043
NaSrB	3	4.85	2.000	1.036	467	4.79	5.19	0.40	4.79	0.719	0.035	0.017	0.037
RhFeSn	1	4.28	3.000	0.418	1016	4.08	4.46	0.38	4.46	0.645	0.170	0.053	0.328
NiVSn	1	4.15	1.000	0.503	34	4.05	4.39	0.34	4.41	0.639	0.157	0.006	0.441
CoFeGe	1	3.89	3.000	0.385	1085	3.81	4.11	0.30	4.11	0.635	0.093	0.069	0.088
NiVSb	1	4.17	2.000	0.435	421	4.11	4.33	0.22	4.39	0.646	0.060	0.025	0.104
RhFeGe	1	4.09	3.000	0.410	1040	4.09	4.29	0.20	4.29	0.633	0.137	0.061	0.245
MnVAs	1	3.97	1.000	0.826	441	3.91	4.07	0.16	4.09	0.667	0.020	0.007	0.044

compounds 111 compounds are half-metals or near-half-metals (these compounds are included in Table 4) and 36 compounds are neither half-metals nor near-half-metals (these compounds are included in Table 5). (For definition of half-metals and near-half-metals, see Section 2.3.)

In the second and fifth columns of Table 4 and 5, we denote the cubic crystal structure with the lowest total energy (ground state) and the cubic crystal structure with the second lowest total energy. The difference, $E_1 - E_2$, between the total energy of the ground state crystal structure, E_1 , and the total energy of the crystal structure with the second lowest total energy, E_2 , is shown in the sixth column of Table 4 and 5. The third and fourth columns of Table 4 and 5 show the lattice constant, a_c , and the total magnetic moment, m_c , calculated for the ground state cubic crystal structure. Calculated values of the bottom edge of the bandgap, E_{max} (we set $E_F = 0$), the bandgap, $E_{\text{gap}} = E_{\text{max}} - E_{\text{min}}$, the spin channel in which the bandgap is located, and the (HM/near-HM) indicator are shown in Table 4 in columns 7–11, respectively.

References to published papers where a specific half-Heusler compound was identified as a half-metal are shown in the last column of Table 4 and 5. Note that the ratio of the number of compounds that we have identified as neither half-metals or near-half-metals (these compounds are listed in Table 5) to the number of compounds that we have identified as half-metals or near-half-metals (these compounds are listed in Table 4) is significantly smaller for half-Heuslers as compared to full-Heuslers (compare the ratio of 36 compounds in Table 5 to the 111 compounds in Table 4 and analogous ratio of 55 compounds in Table 2 to 35 compounds in Table 1). This can be explained by the fact that half-Heusler compounds have significantly larger bandgaps as compared to the full-Heusler compounds (the typical bandgap for half-Heuslers is $\approx 1 \text{ eV}$, while the typical bandgap for full-Heuslers is $\approx 0.3 \text{ eV}$), so the effect of the computational method used or the chosen DFT functional on the conclusion as to whether a given compound is a half-metal or not is much smaller for half-Heuslers as compared to full-Heuslers, so leading to a better agreement between our calculations and previous papers.

The larger values of the bandgaps for half-Heuslers as compared to full-Heuslers can be explained by presence of the *s*-character alkali or alkaline earth elements in most of the considered half-Heusler compounds. The bands with large contributions from *s*-character are typically wider (and, therefore, bandgaps are also wider) as compared to less dispersive *p*- or *d*-character bands. Note that half-Heusler compounds without alkali or alkaline earth elements have smaller bandgaps (see Table 6 and 7).

Analogous to the case of full-Heuslers, some of the compounds in Table 5 have a bandgap in one spin channel, but either with E_{min} located too far above E_F , with $E_{min} - E_F > 0.1$ eV, or E_{max} located too far below E_F , with $E_F - E_{max} > 0.1$ eV. Also, some of the compounds in Table 5 do not have the bandgap but have a very small density of states in one of the spin channels at or near E_F .

Table 6 and 7 present results of calculations of the PMA and half-metallic properties of 100 half-metals and near-half-metals from Table 4 when these compounds undergo a tetragonal distortion (the list of compounds in Table 6 and 7 is that of Table 4 with the exclusion of 10 near-half-metals that do not become half-metals under any tetragonal distortion, and CoTiSb that is a nonmagnetic semiconductor).

Table 6 shows the ground state crystal structure indicator (1, 2, or 3), the lattice constant, a_c , the total magnetic moment,

ADVANCED SCIENCE NEWS _____ www.advancedsciencenews.com



Table 7. Same as Table 6 but for compounds that do not have PMA ($K_v < 0$). Also, we have not calculated T_C for these compounds, so corresponding column is absent in this table.

	Ground state	a _c [Å]	m _c [μ _B]	E _{gap} [eV]	a _{min} [Å]	a _{max} [Å]	w _{hm} [Å]	a _{opt} [Å]	$c'_{\rm opt}$	$K_{\rm mc}$ [meV f.u. ⁻¹]	$K_{\rm sh} \ [{\rm meV f.u.}^{-1}]$	$K_{\rm v} [{\rm MJ} {\rm m}^{-3}]$
KCaC	3	4.68	1.000	1.134	4.08	5.28	1.20	5.10	0.636	0.004	0.004	0.000
NaKAs	3	5.35	1.000	1.605	4.75	5.95	1.20	5.35	0.707	0.000	0.003	-0.005
NaCaC	3	4.36	1.000	1.759	3.76	4.96	1.20	4.46	0.681	0.001	0.006	-0.012
CsCrSe	2	5.14	5.000	2.551	4.54	5.74	1.20	5.54	0.645	0.050	0.077	-0.039
NaKN	3	4.53	1.000	1.828	3.93	5.13	1.20	4.47	0.736	0.001	0.005	-0.010
NaKSb	3	5.65	1.000	1.652	5.17	6.35	1.18	5.61	0.717	0.002	0.003	-0.001
RbVTe	2	5.15	4.000	2.813	4.59	5.73	1.14	4.85	0.777	0.005	0.061	-0.101
LiCaC	3	4.15	1.000	2.166	3.65	4.75	1.10	4.15	0.707	0.000	0.007	-0.021
CsCrTe	2	5.37	5.000	2.263	4.81	5.89	1.08	5.37	0.707	0.000	0.077	-0.113
NaVTe	3	4.84	4.000	3.237	4.28	5.30	1.02	5.28	0.595	0.061	0.062	-0.001
CsVSe	2	5.06	4.000	2.654	4.80	5.70	0.90	5.48	0.637	0.038	0.051	-0.021
RbNbTe	2	5.25	4.000	2.921	5.09	5.91	0.82	5.43	0.669	0.050	0.050	-0.001
KCaB	3	4.98	2.000	0.872	4.76	5.58	0.82	4.76	0.750	0.004	0.017	-0.025
CrScSb	1	4.56	4.000	0.975	4.30	5.12	0.82	4.68	0.669	0.013	0.079	-0.153
MgCaB	3	4.43	1.000	0.765	4.11	4.89	0.78	4.53	0.686	0.003	0.005	-0.006
LiVGe	3	4.19	2.000	0.779	4.13	4.85	0.72	4.85	0.546	0.017	0.022	-0.012
RbCrS	3	4.97	5.000	2.437	4.37	5.09	0.72	4.37	0.880	0.009	0.115	-0.231
FeMnAs	1	3.90	2.000	1.085	3.60	4.26	0.66	4.04	0.661	0.020	0.031	-0.040
RhVSb	1	4.30	1.000	0.859	3.92	4.56	0.64	4.24	0.728	0.002	0.006	-0.012
FeMnSb	1	4.13	2.000	0.984	3.63	4.27	0.64	4.13	0.707	0.000	0.027	-0.087
CsBaGe	3	5.94	1.000	0.509	5.96	6.58	0.62	5.96	0.710	-0.010	0.002	-0.013
CoMnAs	1	3.91	3.000	1.210	3.51	4.13	0.62	3.79	0.756	0.001	0.074	-0.283
CsCrS	3	5.19	5.000	2.032	4.67	5.27	0.60	4.69	0.858	0.046	0.095	-0.089
YMnGe	3	4.58	4.000	0.314	4.54	5.14	0.60	4.68	0.683	0.006	0.077	-0.163
CoVAs	1	3.92	1.000	1.243	3.78	4.36	0.58	3.88	0.719	0.002	0.008	-0.023
CoMnSb	1	4.11	3.000	0.923	3.61	4.17	0.56	4.03	0.735	0.006	0.063	-0.190
KCrS	3	4.79	5.000	2.927	4.19	4.73	0.54	4.93	0.674	0.012	0.104	-0.183
NiCrSi	1	3.85	2.000	0.877	3.47	4.01	0.54	3.81	0.725	0.001	0.034	-0.130
CsNbTe	2	5.36	3.997	2.406	5.50	6.02	0.52	5.56	0.674	0.036	0.047	-0.015
NiMnAs	1	3.99	4.000	0.713	3.63	4.13	0.50	4.13	0.653	0.034	0.117	-0.290
NiMnSb	1	4.18	4.000	0.490	3.80	4.26	0.46	3.80	0.871	0.062	0.113	-0.170
YMnSi	3	4.52	3.981	0.229	4.66	5.08	0.42	4.66	0.675	0.017	0.079	-0.145
CoCrSi	1	3.80	1.000	0.841	3.82	4.20	0.38	3.82	0.701	-0.005	0.009	-0.056
NaCaB	3	4.63	2.000	1.103	4.51	4.83	0.32	4.83	0.673	0.002	0.018	-0.033
NiCrGe	1	3.93	2.000	0.674	3.67	3.99	0.32	3.87	0.731	0.004	0.032	-0.105
RuMnAs	1	4.08	2.000	0.763	3.80	4.10	0.30	4.10	0.697	0.015	0.028	-0.044
NiCrAs	3	3.97	3.000	0.593	3.75	4.03	0.28	4.03	0.686	0.009	0.068	-0.209
FeCrSe	1	3.96	2.000	0.644	3.66	3.94	0.28	3.66	0.826	0.006	0.033	-0.108
NiFeGa	1	3.93	3.000	0.558	3.81	4.09	0.28	4.09	0.647	0.032	0.068	-0.132
LiCaB	3	4.40	2.000	1.163	4.34	4.48	0.14	4.46	0.694	0.001	0.022	-0.054
MnVSb	1	4.20	1.000	0.849	4.08	4.20	0.12	4.26	0.693	0.006	0.006	0.000

 m_c , the bandgap, E_{gap} , and the Curie temperature, T_C , calculated for the ground state cubic structure. Table 6 also shows the minimum, a_{\min} , and the maximum, a_{\max} , values of the in-plane lattice constant for which the tetragonally distorted compound

remains half-metallic (note that for near-half-metals either $a_{\min} > a_c$ or $a_{\max} < a_c$). The values of some a_{\min} and a_{\max} are shown in bold. The bold font means that the true value of a_{\min} is smaller and the true value of a_{\max} is larger than the values



shown in Table 6. The reason for this is that we have not calculated tetragonal distortions which correspond to a too large deviation from the cubic structure (typically, we stopped further calculations when $a_c - a_{min}$ or $a_{max} - a_c$ exceeded 0.6 Å). Thus, the width of the half-metallic range, $w_{hm} = a_{max} - a_{min}$, that is presented in Table 6 is a lower bound of the true w_{hm} for the compounds that have a_{min} or a_{max} shown in bold font. Note that due to larger values of the bandgaps w_{hm} is significantly wider for the half-Heuslers as compared to the full-Heuslers. As we discussed above, larger calculated values of E_{gap} and w_{hm} make the half-metallic properties of compounds more stable and less susceptible to changes in the in-plane and out-of-plane lattice constants, details of the computational method, chosen DFT functional, finite temperature effects, and effects of defects such as impurities and dislocations.

The values of a_{opt} , shown in Table 6, are the in-plane lattice constant that provides the largest (optimal) MCA energy, K_{mc} , for *a* in the half-metallicity range ($a_{\min} \leq a_{opt} \leq a_{\max}$). The values of c'_{opt} are the out-of-plane dimensionless constant that corresponds to a_{opt} . We found that due to the wide width of the half-metallic range of typical half-metallic half-Heusler compounds, K_{mc} as a function of a in the range $a_{\min} \le a \le a_{\max}$ often does not follow a simple linear function (as opposed to the case of full-Heuslers), while it still crosses zero at $a = a_c$ due to the cubic symmetry at this value. Therefore, a_{opt} in Table 6 often do not coincide with a_{\min} or a_{\max} . The values of K_{\min} , K_{sh} , and K_v that correspond to the optimal values of the lattice constants a_{opt} and c'_{opt} are shown in Table 6. Note that in Table 6 we show only the compounds with PMA ($K_v > 0$). Thus, the compounds shown in Table 6 are compounds that can simultaneously be half-metals and have PMA under an optimal tetragonal distortion.

In order to understand the origin of the large Curie temperature for half-Heusler compounds presented in Table 6, we studied the contribution to the $J^{\mu\nu}$ matrix of exchange constants J_{ij} for various sites *i* and *j* for compounds with $T_{\rm C} > 400$ K. We found that for compounds XYZ where both X and Y elements are the alkali or alkaline earth elements the main contribution to $J^{\mu\nu}$ comes from J_{ij} with *i* and *j* being the nearest-neighbor Z (main group element) sites. For compounds XYZ where X is alkali or alkaline earth element and Y is the transitional metal, the main contribution to $J^{\mu\nu}$ comes from J_{ij} with *i* and *j* being the nearestneighbor Y (transitional metal) sites. For compounds XYZ where both X and Y are transitional metals, the main contribution to $J^{\mu\nu}$ comes from J_{ij} with *i* and *j* being the nearest-neighbor transitional metal sites (X and Y).

Table 7 contains the same information as Table 6 (except that we have not calculated the Curie temperature) but for compounds with $K_v \leq 0$ at the optimal values of the lattice constants $a_{\rm opt}$ and $c'_{\rm opt}$. Thus, compounds shown in Table 7 cannot simultaneously be half-metals and have PMA even under optimal tetragonal distortions. Note that the compounds listed in Table 6 and 7 are ordered by the values of the width of the half-metallicity range, $w_{\rm hm}$.

Twenty compounds from Table 6, KCrTe, NaCsP, NaCsAs, CsRbAs, KCaGe, CsBaC, LiSrGe, KCaSn, NaCaGe, KTaSn, RbTaGe, CsSrSn, CsSrC, RbTaSi, RbNbSi, CoCrAs, NaCaSn, RhCrGe, LiCaGe, and RhFeSn, have a wide half-metallicity range, $w_{\rm hm} \ge 0.40$ Å, a large bandgap, $E_{\rm gap} \ge 0.40$ eV, and a

relatively large PMA under the optimal tetragonal distortion, $K_{\rm v} \ge 0.25 {\rm MJ m^{-3}}$. Seven of these compounds, CsBaC, RbTaGe, CsSrC, RbTaSi, RbNbSi, CoCrAs, and RhFeSn, also have a large $T_{\rm C} > 410 {\rm K}$. These seven compounds form our list of the most promising half-Heusler compounds for MTJ devices.

4. Conclusion

In this work, we have proposed a mechanism by which a Heusler compound can simultaneously display both PMA and halfmetallic properties, which is highly desirable for STT-MRAM devices. In particular, we propose that a tetragonal distortion of a thin film of a half-metallic cubic full-Heusler or half-Heusler compound can be set in the film by having the film adopt the in-plane lattice constant of an underlayer material during the thin film deposition process. The value of the distortion can be tuned by using an underlayer material with a suitable in-plane lattice constant. The optimal distortion should be large enough to make the Heusler compound tetragonal with sufficient PMA, while, simultaneously, small enough to maintain the half-metallic properties (and, therefore, high TMR properties) of the compound.

We performed DFT calculations for 90 full-Heuslers that were identified in the literature as half-metals. We found that 24 of them are half metals and that 14 of these half-metals (see Table 3) can simultaneously keep their half-metallicity and have PMA under an optimal tetragonal distortion (optimal in-plane lattice constant). Our most promising list of full-Heusler compounds for MTJ devices includes six compounds, namely, Mn₂CoSi, Mn₂CoAl, Co₂CrSi, Mn₂CuSi, Mn₂CoGe, and Co₂CrGe, that simultaneously have a wide half-metallicity range, $w_{\rm hm} \ge 0.24$ Å, a relatively large bandgap, $E_{\rm gap} \ge 0.30$ eV, a large Curie temperature, $T_{\rm C} \ge 490$ K, and a relatively large PMA under an optimal tetragonal distortion, $K_{\rm v} \ge 0.18$ MJ m⁻³.

We also performed DFT calculations for 147 half-Heuslers that were identified in the literature as half-metals. We found that 94 of them are half metals and that 59 of these half-metals (see Table 6) can simultaneously keep their half-metallicity and have PMA under an optimal tetragonal distortion (optimal in-plane lattice constant). Our most promising list of half-Heusler compounds for MTJ devices includes seven compounds CsBaC, RbTaGe, CsSrC, RbTaSi, RbNbSi, CoCrAs, and RhFeSn that simultaneously display a wide half-metallicity range, $w_{\rm hm} \ge 0.40$ Å, a relatively large bandgap, $E_{\rm gap} \ge 0.40$ eV, a large Curie temperature, $T_{\rm C} \ge 410$ K, and a relatively large PMA under an optimal tetragonal distortion, $K_{\rm v} \ge 0.25$ MJ m⁻³.

Acknowledgements

S.F. acknowledges the CNMS User support of the Oak Ridge National Laboratory Division of Scientific User facilities.

Open Access funding enabled and organized by Projekt DEAL.

Conflict of Interest

The authors declare no conflict of interest.

4DVANCED

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

density functional theory (DFT) calculations, half-metallic, Heusler compounds, magnetic random-access memory (MRAM), perpendicular magnetic anisotropy, tunneling magnetoresistance

> Received: November 8, 2022 Revised: December 29, 2022 Published online:

- [1] J. Åkerman, Science 2005, 308, 508.
- [2] S. Ikeda, K. Miura, H. Yamamoto, K. Mizunuma, H. D. Gan, M. Endo, S. Kanai, J. Hayakawa, F. Matsukura, H. Ohno, *Nat. Mater.* **2010**, *9*, 721.
- [3] H. Sato, E. C. I. Enobio, M. Yamanouchi, S. Ikeda, S. Fukami, S. Kanai, F. Matsukura, H. Ohno, *Appl. Phys. Lett.* 2014, 105, 062403.
- [4] S. S. P. Parkin, US Patent 8,008,097, 2003.
- [5] S. S. P. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, S.-H. Yang, *Nat. Mater.* 2004, *3*, 862.
- [6] T. Graf, C. Felser, S. S. P. Parkin, Prog. Solid State Chem. 2011, 39, 1.
- [7] B. Balke, G. H. Fecher, J. Winterlik, C. Felser, Appl. Phys. Lett. 2007, 90, 152504.
- [8] H. Kurt, N. Baadji, K. Rode, M. Venkatesan, P. Stamenov, S. Sanvito, J. M. D. Coey, *Appl. Phys. Lett.* **2012**, *101*, 132410.
- [9] A. Sugihara, S. Mizukami, Y. Yamada, K. Koike, T. Miyazaki, Appl. Phys. Lett. 2014, 104, 132404.
- [10] S. Mizukami, A. Sakuma, A. Sugihara, T. Kubota, Y. Kondo, H. Tsuchiura, T. Miyazaki, Appl. Phys. Express 2013, 6, 123002.
- [11] M. Li, X. Jiang, M. G. Samant, C. Felser, S. S. P. Parkin, Appl. Phys. Lett. 2013, 103, 032410.
- [12] A. Köhler, I. Knez, D. Ebke, C. Felser, S. S. P. Parkin, Appl. Phys. Lett. 2013, 103, 162406.
- [13] J. Jeong, Y. Ferrante, S. V. Faleev, M. G. Samant, C. Felser, S. S. P. Parkin, *Nat. Commun.* **2016**, *7*, 10276.
- [14] P. C. Filippou, J. Jeong, Y. Ferrante, S.-H. Yang, T. Topuria, M. G. Samant, S. S. P. Parkin, *Nat. Commun.* 2018, 9, 4653.
- [15] S. Faleev, Y. Ferrante, J. Jeong, M. G. Samant, B. Jones, S. S. P. Parkin, *Phys. Rev. Mater.* 2017, 1, 024402.
- [16] H.-X. Liu, Y. Honda, T. Taira, K.-I. Matsuda, M. Arita, T. Uemura, M. Yamamoto, Appl. Phys. Lett. 2012, 101, 132418.
- [17] M. Jourdan, J. Minár, J. Braun, A. Kronenberg, S. Chadov, B. Balke, A. Gloskovskii, M. Kolbe, H. J. Elmers, G. Schönhense, H. Ebert, C. Felser, M. Kläui, *Nat. Commun.* **2014**, *5*, 3974.
- [18] G. Kresse, J. Furthmüller, Phys. Rev. B 1996, 54, 11169.
- [19] P. E. Blöchl, Phys. Rev. B 1994, 50, 17953.
- [20] G. Kresse, D. Joubert, Phys. Rev. B 1999, 59, 1758.
- [21] J. P. Perdew, K. Burke, M. Ernzerhof, Phys. Rev. Lett. 1996, 77, 3865.
- [22] A. I. Liechtenstein, M. I. Katsnelson, V. P. Antropov, V. A. Gubanov, J. Magn. Magn. Mater. 1987, 67, 65.
- [23] P. W. Anderson, in *Solid State Physics*, Vol. 14 (Eds: F. Seitz, D. Turnbull), Academic Press, New York **1955**, pp. 99–214.
- [24] O. K. Andersen, O. Jepsen, Phys. Rev. Lett. 1984, 53, 2571.



- [25] M. van Schilfgaarde, W. R. L. Lambrecht, in *Tight-Binding Approach to Computational Materials Science* (Eds: L. Colombo, A. Gonis, P. Turchi), MRS Symposia Proceedings No. 91, Materials Research Society, Pittsburgh **1998**, pp. 137–142.
- [26] J. Kübler, Physica B+C 1984, 127, 257.
- [27] I. Galanakis, P. H. Dederichs, N. Papanikolaou, Phys. Rev. B 2002, 66, 174429.
- [28] S. V. Faleev, Y. Ferrante, J. Jeong, M. G. Samant, B. Jones, S. S. P. Parkin, *Phys. Rev. B* **2017**, *95*, 045140.
- [29] S. Skaftouros, K. Özdoğan, E. Şaşıoğlu, I. Galanakis, Phys. Rev. B 2013, 87, 024420.
- [30] P. C. Filippou, S. V. Faleev, C. Garg, J. Jeong, Y. Ferrante, T. Topuria, M. G. Samant, S. S. P. Parkin, *Sci. Adv.* 2022, *8*, eabg2469.
- [31] I. Galanakis, P. Mavropoulos, J. Phys.: Condens. Matter 2007, 19, 315213.
- [32] B. Balke, G. H. Fecher, H. C. Kandpal, C. Felser, K. Kobayashi, E. Ikenaga, J.-J. Kim, S. Ueda, *Phys. Rev. B* 2006, *74*, 104405.
- [33] X.-Q. Chen, R. Podloucky, P. Rogl, J. Appl. Phys. 2006, 100, 113901.
- [34] H. C. Kandpal, G. H. Fecher, C. Felser, J. Phys. D: Appl. Phys. 2007, 40, 1507.
- [35] D. P. Rai, A. Shankar, C. Sandeep, M. P. Ghimire, R. K. Thapa, J. Theor. Appl. Phys. 2013, 7, 3.
- [36] R. Y. Umetsu, K. Kobayashi, R. Kainuma, A. Fujita, K. Fukamichi, K. Ishida, A. Sakuma, *Apl. Phys. Lett.* **2004**, *85*, 2011.
- [37] R. A. de Groot, F. M. Mueller, P. G. van Engen, K. H. J. Buschow, Phys. Rev. Lett. 1983, 50, 2024.
- [38] I. Galanakis, E. Sasioglu, K. Ozdogan, Phys. Rev. B 2008, 77, 214417.
- [39] I. Galanakis, P. Mavropoulos, P. H. Dederichs, J. Phys. D: Appl. Phys. 2006, 39, 765.
- [40] J. Ma, V. I. Hegde, K. Munira, Y. Xie, S. Keshavarz, D. T. Mildebrath, C. Wolverton, A. W. Ghosh, W. H. Butler, *Phys. Rev. B* 2017, 95, 024411.
- [41] M. Safavi, M. Moradi, M. Rostami, J. Supercond. Novel Magn. 2017, 30, 989.
- [42] H. C. Kandpal, C. Felser, R. Seshadri, J. Phys. D: Appl. Phys. 2006, 39, 776.
- [43] K. Sedeek, H. Handour, N. Makram, S. A. Said, J. Magn. Magn. Mater. 2016, 407, 218.
- [44] X. Wang, Z. Cheng, G. Liu, Materials 2017, 10, 1078.
- [45] M. A. Sattar, S. A. Ahmad, F. Hussain, C. Cazorla, J. Materiomics 2019, 5, 404.
- [46] M. Rostami, Surf. Sci. 2018, 674, 103.
- [47] A. Lakdja, H. Rozale, A. Sayede, A. Chahed, J. Magn. Magn. Mater. 2014, 354, 235.
- [48] R. Umamaheswari, D. Vijayalakshmi, G. Kalpana, *Physica B* 2014, 448, 256.
- [49] A. Abada, N. Marbouh, J. Supercond. Novel Magn. 2020, 33, 889.
- [50] J. M. Khalaf Al-Zyadi, M. H. Jolan, K.-L. Yao, J. Magn. Magn. Mater. 2016, 403, 8.
- [51] J. Chen, G. Y. Gao, K. L. Yao, M. H. Song, J. Alloys Compd. 2011, 509, 10172.
- [52] J.-S. Zhao, Q. Gao, L. Li, H.-H. Xie, X.-R. Hu, C.-L. Xu, J.-B. Deng, Intermetallics 2017, 89, 65.
- [53] I. Galanakis, K. Özdoğan, E. Şaşıoğlu, J. Appl. Phys. 2008, 104, 083916.
- [54] H. Rozale, A. Amar, A. Lakdja, A. Moukadem, A. Chahed, J. Magn. Magn. Mater. 2013, 336, 83.
- [55] H. Rozale, M. Khetir, A. Amar, A. Lakdja, A. Sayede, O. Benhelal, Superlattices Microstruct. 2014, 74, 146.
- [56] A. Lakdja, H. Rozale, A. Chahed, O. Benhelal, J. Alloys Compd. 2013, 564, 8.
- [57] J. G. Azadani, K. Munira, J. Romero, J. Ma, C. Sivakumar, A. W. Ghosh, W. H. Butler, J. Appl. Phys. 2016, 119, 043904.

4DVANCED



- [58] M. A. Sattar, M. Rashid, F. Hussain, M. Imran, M. R. Hashmi, A. Laref, S. A. Ahmad, Solid State Commun. 2018, 278, 10.
- [59] R. L. Zhang, L. Damewood, C. Y. Fong, L. H. Yang, R. W. Peng, C. Felser, AIP Adv. 2016, 6, 115209.
- [60] L. Damewood, B. Busemeyer, M. Shaughnessy, C. Y. Fong,
 L. H. Yang, C. Felser, *Phys. Rev. B* 2015, *91*, 064409.
- [61] W. Huang, X. Wang, X. Chen, W. Lu, L. Damewood, C. Y. Fong, J. Magn. Magn. Mater. 2015, 377, 252.