

Heusler compounds with perpendicular magnetic anisotropy and large tunneling magnetoresistance

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In the present work we suggest a recipe for finding tetragonal Heusler compounds with perpendicular magnetic anisotropy (PMA) that also exhibit large tunneling magnetoresistance (TMR) when used as electrodes in magnetic tunnel junction devices with suitable tunneling barrier materials. We performed density-functional theory calculations for 286 Heusler compounds and identified 116 stable tetragonal compounds. Ten of these compounds are predicted to have strong PMA and, simultaneously, exponentially increasing TMR with increasing tunneling barrier thickness due to the so-called Brillouin zone spin filtering effect. Experimental measurements performed for 25 Heusler compounds theoretically identified as tetragonal show that ten of these compounds indeed have tetragonal structure with PMA.

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I. INTRODUCTION

Key to the successful development of spin-transfer torque magnetic random access memory (STT-MRAM), one of the most promising emerging nonvolatile memory technologies today, are new magnetic materials for magnetic tunnel junction (MTJ) memory elements that have sufficient stability against thermal fluctuations to sustain deeply scaled devices. The magnetic electrodes must possess sufficient perpendicular magnetic anisotropy (PMA) that their magnetizations lie perpendicular to the plane of the MTJ device, since this allows for reduced currents to switch the magnetization of the electrode that forms the memory layer of the device using spin torque [1,2]. The most promising magnetic materials to date are considered to be alloys formed from Co, Fe, and B, in conjunction with MgO tunnel barriers [1,3,4]. Unfortunately, PMA of CoFeB layers arises from the interfaces between these layers and the tunnel barrier and/or underlayer and is too weak to overcome thermal fluctuations when the device has a critical dimension $\lesssim 20$ nm.

Magnetic materials in which the PMA is derived from volume magnetocrystalline anisotropy (MCA) are then needed. One of the most promising classes of such materials is the Heusler alloys—compounds having the chemical formula X_2YZ wherein X and Y are transition metals, or lanthanides (rare-earth metals), and Z is the main group element [5]. While the parent Heusler compounds are cubic and do not exhibit magnetic anisotropy, the structure of some of these compounds is found to be tetragonally distorted and thus could potentially have large PMA.

Some examples of tetragonal Heusler compounds are $Mn_{3-x}Ga$ [6] and Mn_3Ge [7]. Thin films of these materials have been shown to exhibit large PMA for films grown epitaxially on single crystalline substrates such as $SrTiO_3(001)$ or $MgO(001)$ [7–11] and on amorphous substrates [$Si(001)/SiO_2$] [12]. Unfortunately, the experimental values of the tunneling magnetoresistance (TMR) for MTJs with $Mn_{3-x}Ga$ or Mn_3Ge

electrodes and MgO spacer were found to be very small, far below the application range [12].

The goal of the present paper is to identify promising tetragonal Heusler compounds that possess PMA and exhibit high TMR either due to large spin polarization of bulk Heusler compound or due to the so-called Brillouin zone (BZ) spin filtering effect [13] for the MTJ with suitable spacer. Realization of the high TMR in a MTJ system with electrodes that have low crystal symmetry where PMA could be simultaneously achieved is of significant technological interest for spintronics applications, and, in particular, in the context of novel STT-MRAM technology that has a potential to become a “universal memory” [14] combining all the strengths and none of the weaknesses of existing memory types.

II. THEORETICAL SEARCH FOR TETRAGONAL HEUSLER COMPOUNDS WITH PMA AND LARGE TMR

A. Crystal structure

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

Regular [Fig. 1(b)] and inverse [Fig. 1(d)] tetragonal Heusler structures can be obtained from regular and inverse cubic structures, correspondingly, by stretching (or compressing) parent cubic structure along the z axis. Tetragonal unit cells shown in Figs. 1(b) and 1(d) are rotated 45° around the z axis relative to the parent cubic structures shown in Figs. 1(a) and 1(c), correspondingly. [Note that only part of atoms from

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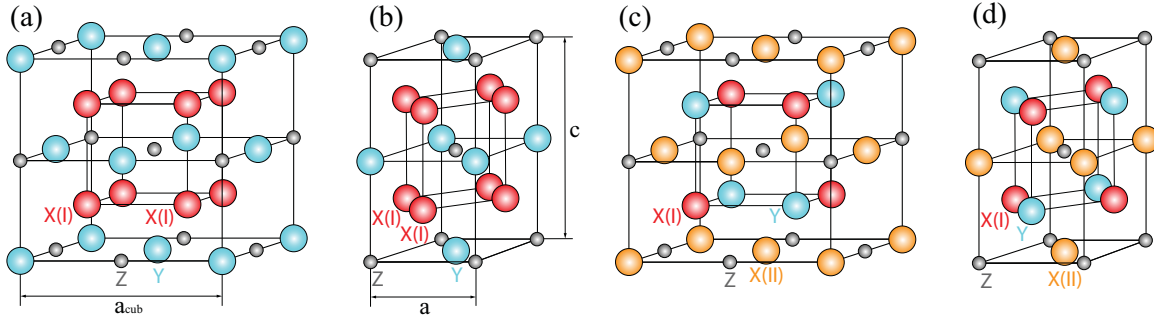


FIG. 1. (a) Regular and (c) inverse cubic Heusler structure. (b) Regular and (d) inverse tetragonal Heusler structure.

Figs. 1(a) and 1(c) are shown in Figs. 1(b) and 1(d).] Lattice constant a_{cub} of the cubic Heusler is shown in Fig. 1(a) and lattice constants a and c of the tetragonal Heusler are shown in Fig. 1(b). For characterization of the tetragonal unit cell we use dimensionless parameter $c' = c/(2a)$ that is equal to $1/\sqrt{2}$ for the cubic structure, and vary between 0.8 and 1.1 for most of the tetragonal Heuslers we found (see Table I). Note that for $c' = 1$ tetragonal structure would become the fcc structure if all four atoms of the compound could be considered as equivalent.

B. Computational details

We performed density functional theory (DFT) calculations for both the regular and inverse structures (with various magnetic configurations) of 286 Heusler compounds [15] using the VASP program [16] with projector augmented wave (PAW) potentials [17,18] and Perdew-Burke-Ernzerhof (PBE) GGA/DFT functional [19]. In particular, we performed calculations for Heusler compounds X_2YZ with $X = \{\text{Mn, Fe, Co}\}$ and $YZ = \{\text{Mn, Fe, Co, Ni, Cu}\}\{\text{Al, Ga, Si, Ge, Sn, Sb}\}$, and $YZ = \{\text{Mo, Ru, Rh, Pd, W, Os, Ir, Pt}\}\{\text{Ga, In, Ge, Sn, Sb}\}$, $X = \{\text{Ru, Rh, Pd}\}$ and $YZ = \{\text{Mn, Fe, Co}\}\{\text{Ga, In, Ge, Sn, Sb}\}$, $X = \text{Ni}$ and $YZ = \{\text{Mn, Fe, Co}\}\{\text{Al, Ga, Si, Ge, Sn, Sb}\}$, $X = \text{Mn}$ and $YZ = \{\text{Fe, Co, Ni, Cu}\}\{\text{In}\}$, and compounds X_3Z with $X = \{\text{Mn, Fe, Co}\}$ and $Z = \{\text{In, P, As}\}$. For binary compounds X_3Z we also considered hexagonal structures with X atoms forming a kagome lattice in a plane with a Z atom in the center of the hexagon (see, e.g., Ref. [20] for figure of the hexagonal structure).

The results of calculations are summarized in Table I for 116 compounds with tetragonal lowest energy configuration and $E_{21} \geq 0.05$ eV, where phase stability energy, E_{21} , is defined as the difference between the total energy of the second lowest energy configuration and the total energy of the lowest energy configuration. The remaining 170 compounds that are not included in Table I either have cubic or hexagonal lowest energy configuration or tetragonal lowest energy configuration with low phase stability energy, $E_{21} < 0.05$ eV. (See Ref. [15] for explanation of such a large share of stable tetragonal compounds—116—out of 286 studied compounds.) The convergence of presented results was verified by varying the number of divisions in reciprocal space from $10 \times 10 \times 10$ to $18 \times 18 \times 18$ and the energy cutoff from 400 to 520 eV.

The MCA energy, K_{mc} , of tetragonal Heusler compounds is calculated as the difference between total energies of

states with magnetization along the x axis and the z axis, $K_{mc} = E_{(100)} - E_{(001)}$, where positive K_{mc} means out-of-plane magnetization. We also calculated the volume magnetic anisotropy, $K_v = K_{mc} - K_{sh}$, where $K_{sh} = \mu_0 M_s^2 V/2$ is the shape anisotropy energy of thin film per unit cell of volume V , M_s is saturation magnetization, and μ_0 is vacuum permeability.

We calculated Curie temperature, T_C , within the standard mean-field approximation [21] using the exchange constants, J_{ij} , of the effective Heisenberg Hamiltonian (i and j are the site indexes). In this approach T_C can be estimated as $k_B T_C = 2/3 J_{\text{max}}$, where J_{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 the fixed index in sublattice μ and the sum is taken over sites in sublattice ν . The exchange constants J_{ij} were calculated by using the Green's function approach implemented within the linear muffin-tin orbitals and atomic spheres approximation (LMTO-ASA) framework [22,23].

For each stable tetragonal compound Table I shows the lattice parameters a and c' ; the lowest and the second lowest energy configurations labels s_1 and s_2 ; magnetic moment m ; phase stability energy E_{21} ; total spin polarization SP_t as well as the spin polarization of individual termination layers SP_1 and SP_2 ; anisotropy constants K_{mc} , K_{sh} , and K_v ; and T_C . For ternary compounds labels $s_1, s_2 = tr, ti, cr, ci$ represent tetragonal regular, tetragonal inverse, cubic regular, or cubic inverse phase, correspondingly. For binary compounds $s_1, s_2 = t, c, h$ represent tetragonal, cubic, or hexagonal phase, correspondingly. Since only stable tetragonal compounds are presented in Table I, s_1 always indicates tetragonal phase, $s_1 = tr, ti, cr, ci$. Anisotropy constants K_{mc} and K_{sh} are shown in Table I in $\frac{\text{meV}}{\text{f.u.}}$ units, while $K_v = K_{mc} - K_{sh}$ is converted from $\frac{\text{meV}}{\text{f.u.}}$ to $\frac{\text{MJ}}{\text{m}^3}$ units. Positive K_v indicates that this compound has PMA. Three values of the spin polarization shown in Table I— SP_t , SP_1 , and SP_2 —are defined as $\frac{\text{DOS}_1 - \text{DOS}_2}{\text{DOS}_1 + \text{DOS}_2}$, where $\text{DOS}_{1/2}$ is density of states at the Fermi energy, E_F , of majority/minority electrons calculated for the whole system, projected to a two-atom termination layer (xy plane) that does not contain atom Z , or to a two-atom termination layer that contains atom Z , correspondingly.

C. Compounds with expected large TMR due to Brillouin zone spin filtering effect

The so-called BZ spin filtering effect [13] in a ME/MgO/ME MTJ system occurs if the magnetic electrode (ME) has states at the Fermi energy, E_F , along the Γ - Z line in

TABLE I. Calculated lattice parameters, a and c' ; labels of the lowest energy configuration, s_1 , and the second lowest energy configuration, s_2 ; the phase stability energy, E_{21} ; magnetic moment, m ; total spin polarization, SP_t ; spin polarizations, SP_1 and SP_2 , of two termination layers; anisotropy constants, K_{mc} , K_{sh} , and K_v ; and the Curie temperature, T_C , for 116 stable tetragonal Heuslers with $E_{21} \geq 0.05$ eV (see text for details).

	a (Å)	c'	s_1	s_2	E_{21} (eV)	m (μ_B)	SP_t	SP_1	SP_2	K_{mc} $\frac{\text{meV}}{\text{f.u.}}$	K_{sh} $\frac{\text{meV}}{\text{f.u.}}$	K_v $\frac{\text{MJ}}{\text{m}^3}$	T_C (K)
Mn ₂ NiAl	3.81	0.88	<i>ti</i>	<i>ci</i>	0.06	1.01	0.27	0.36	0.03	0.167	0.007	0.53	887
Mn ₃ Ga	3.78	0.94	<i>t</i>	<i>c</i>	0.19	1.79	-0.60	-0.67	-0.49	0.884	0.021	2.72	658
Mn ₂ FeGa	3.69	0.98	<i>ti</i>	<i>ci</i>	0.14	0.84	-0.58	-0.71	-0.33	0.494	0.005	1.59	751
Mn ₂ NiGa	3.79	0.91	<i>ti</i>	<i>ci</i>	0.12	0.99	0.66	0.70	0.54	0.162	0.007	0.51	821
Mn ₃ In	3.95	0.96	<i>t</i>	<i>h</i>	0.15	1.93	-0.72	-0.81	-0.53	1.218	0.021	3.24	461
Mn ₂ FeIn	3.89	0.98	<i>ti</i>	<i>ti</i>	0.18	0.94	-0.75	-0.63	-0.86	0.659	0.005	1.82	715
Mn ₂ CoIn	3.90	0.96	<i>ti</i>	<i>ci</i>	0.06	0.08	0.65	0.76	0.30	0.122	0.000	0.34	170
Mn ₂ NiSi	3.61	0.97	<i>ti</i>	<i>ci</i>	0.10	0.32	0.06	0.19	-0.18	0.140	0.001	0.49	548
Mn ₂ CuSi	3.71	0.94	<i>tr</i>	<i>ci</i>	0.19	4.92	-0.29	-0.29	-0.14	0.474	0.170	1.02	426
Mn ₃ Ge	3.73	0.95	<i>t</i>	<i>c</i>	0.10	1.01	-0.72	-0.77	-0.63	0.770	0.007	2.48	503
Mn ₂ CoGe	3.74	0.92	<i>ti</i>	<i>ci</i>	0.07	0.97	0.78	0.85	0.50	0.208	0.007	0.67	665
Mn ₂ NiGe	3.73	0.95	<i>ti</i>	<i>ci</i>	0.08	0.31	-0.26	-0.17	-0.47	0.135	0.001	0.44	744
Mn ₂ CuGe	3.77	0.95	<i>tr</i>	<i>ci</i>	0.21	5.28	-0.31	-0.30	-0.17	0.646	0.185	1.45	397
Mn ₃ Sn	3.93	0.95	<i>t</i>	<i>h</i>	0.17	1.04	-0.87	-0.92	-0.68	0.958	0.006	2.65	264
Mn ₂ CoSn	3.96	0.90	<i>ti</i>	<i>ci</i>	0.15	1.00	0.89	0.93	0.69	0.337	0.006	0.95	603
Mn ₂ CuSn	3.91	0.98	<i>tr</i>	<i>ci</i>	0.06	5.73	-0.30	-0.32	-0.06	0.965	0.189	2.12	421
Mn ₃ Sb	3.82	1.02	<i>t</i>	<i>h</i>	0.14	1.14	0.23	0.09	0.58	0.241	0.008	0.66	284
Mn ₂ NiSb	3.81	1.02	<i>tr</i>	<i>ti</i>	0.11	5.85	-0.47	-0.42	-0.51	0.926	0.204	2.05	132
Mn ₂ CuSb	3.86	1.01	<i>tr</i>	<i>ti</i>	0.27	5.45	-0.33	-0.36	-0.05	0.963	0.172	2.18	53
Mn ₂ RuGa	3.81	0.97	<i>ti</i>	<i>ci</i>	0.10	0.26	0.03	-0.14	0.26	0.599	0.000	1.79	1107
Mn ₂ RhGa	3.83	0.97	<i>ti</i>	<i>ci</i>	0.17	0.07	0.65	0.81	0.26	0.302	0.000	0.89	1112
Mn ₂ PdGa	3.93	0.92	<i>ti</i>	<i>ci</i>	0.10	0.92	0.46	0.63	0.00	0.021	0.005	0.05	551
Mn ₂ MoIn	4.09	0.88	<i>tr</i>	<i>cr</i>	0.09	3.73	0.26	0.20	0.60	1.473	0.078	3.71	477
Mn ₂ RuIn	3.94	1.00	<i>ti</i>	<i>tr</i>	0.21	0.30	0.17	-0.03	0.45	0.865	0.000	2.26	992
Mn ₂ RhIn	3.98	0.98	<i>ti</i>	<i>ci</i>	0.22	0.05	0.59	0.80	0.19	-0.002	0.000	0.00	825
Mn ₂ RhGe	3.85	0.94	<i>ti</i>	<i>ci</i>	0.22	0.77	-0.07	0.23	-0.66	0.019	0.004	0.04	463
Mn ₂ MoSn	3.86	1.08	<i>ti</i>	<i>tr</i>	0.07	0.14	-0.12	0.02	-0.31	-0.473	0.000	-1.22	768
Mn ₂ RuSn	3.97	0.97	<i>ti</i>	<i>ci</i>	0.21	0.00	0.91	0.96	0.77	0.431	0.000	1.14	783
Mn ₂ RhSn	4.03	0.93	<i>ti</i>	<i>ci</i>	0.14	0.75	-0.45	-0.20	-0.83	-0.068	0.003	-0.19	309
Mn ₂ PdSn	4.17	0.89	<i>ti</i>	<i>ci</i>	0.05	0.20	-0.04	0.31	-0.55	-0.074	0.000	-0.19	710
Mn ₂ MoSb	3.88	1.06	<i>ti</i>	<i>ci</i>	0.26	0.20	0.47	0.43	0.60	0.029	0.000	0.08	647
Mn ₂ RuSb	3.94	0.98	<i>ti</i>	<i>ci</i>	0.17	0.17	-0.38	-0.19	-0.74	-0.208	0.000	-0.56	321
Mn ₂ RhSb	4.00	0.96	<i>ti</i>	<i>tr</i>	0.16	0.00	0.45	0.51	0.13	0.106	0.000	0.28	747
Mn ₂ OsGa	3.80	0.98	<i>ti</i>	<i>ci</i>	0.15	0.28	0.08	0.00	0.20	3.291	0.000	9.81	929
Mn ₂ IrGa	3.83	0.97	<i>ti</i>	<i>ci</i>	0.27	0.07	0.57	0.73	0.23	2.760	0.000	8.11	962
Mn ₂ PtGa	3.90	0.94	<i>ti</i>	<i>tr</i>	0.11	0.87	0.34	0.54	-0.23	-0.210	0.005	-0.62	375
Mn ₂ OsIn	3.92	1.01	<i>ti</i>	<i>tr</i>	0.27	0.29	0.23	0.11	0.39	4.699	0.000	12.37	734
Mn ₂ IrIn	3.96	0.99	<i>ti</i>	<i>tr</i>	0.30	0.04	0.78	0.90	0.43	1.754	0.000	4.57	497
Mn ₂ OsGe	3.81	0.97	<i>ti</i>	<i>ci</i>	0.06	0.03	-0.91	-0.94	-0.83	2.620	0.000	7.82	476
Mn ₂ IrGe	3.83	0.96	<i>ti</i>	<i>tr</i>	0.25	0.51	-0.62	-0.44	-0.86	0.046	0.002	0.13	365
Mn ₂ OsSn	3.96	0.98	<i>ti</i>	<i>ci</i>	0.33	0.02	-0.91	-0.95	-0.77	4.047	0.000	10.65	611
Mn ₂ IrSn	3.99	0.96	<i>ti</i>	<i>ci</i>	0.22	0.46	-0.65	-0.45	-0.89	-0.578	0.001	-1.52	227
Mn ₂ WSb	3.84	1.08	<i>ti</i>	<i>cr</i>	0.23	0.06	0.49	0.44	0.59	-0.894	0.000	-2.34	603
Mn ₂ OsSb	3.96	0.97	<i>ti</i>	<i>ci</i>	0.24	0.17	-0.46	-0.26	-0.78	0.991	0.000	2.64	206
Fe ₂ CuAl	3.57	1.08	<i>tr</i>	<i>ci</i>	0.16	4.65	-0.11	-0.24	0.07	0.135	0.148	-0.04	1295
Fe ₂ CuGa	3.60	1.06	<i>tr</i>	<i>ti</i>	0.22	4.73	-0.14	-0.33	0.29	0.156	0.153	0.01	1238
Fe ₂ NiSi	3.49	1.06	<i>tr</i>	<i>ci</i>	0.12	4.68	0.20	0.09	0.39	0.248	0.164	0.30	1083
Fe ₂ CuSi	3.55	1.03	<i>tr</i>	<i>ti</i>	0.10	4.24	-0.66	-0.74	-0.46	0.588	0.132	1.59	675
Fe ₂ NiGe	3.54	1.07	<i>tr</i>	<i>ci</i>	0.16	4.83	0.04	-0.14	0.36	0.290	0.166	0.42	1081
Fe ₂ CuGe	3.61	1.05	<i>tr</i>	<i>ti</i>	0.15	4.59	-0.59	-0.68	-0.33	0.288	0.144	0.47	615
Fe ₂ MnSn	3.97	0.90	<i>ti</i>	<i>ci</i>	0.08	7.34	-0.18	-0.23	-0.04	0.773	0.323	1.28	1012
Fe ₂ NiSn	3.68	1.09	<i>tr</i>	<i>ci</i>	0.06	4.97	-0.18	-0.36	0.14	0.347	0.153	0.57	1077
Fe ₂ CuSn	3.76	1.06	<i>tr</i>	<i>ti</i>	0.11	4.87	-0.40	-0.51	0.00	-0.630	0.142	-2.19	849
Fe ₂ NiSb	3.70	1.09	<i>tr</i>	<i>ti</i>	0.13	5.41	-0.20	-0.40	0.13	-0.395	0.179	-1.67	1210

TABLE I. (Continued.)

	a (Å)	c'	s_1	s_2	E_{21} (eV)	m (μ_B)	SP_t	SP_1	SP_2	K_{mc} $\frac{\text{meV}}{\text{f.u.}}$	K_{sh} $\frac{\text{meV}}{\text{f.u.}}$	K_v $\frac{\text{MJ}}{\text{m}^3}$	T_C (K)
Fe ₂ PdGe	3.67	1.09	<i>tr</i>	<i>ti</i>	0.07	5.12	-0.27	-0.46	0.19	0.429	0.164	0.79	1086
Fe ₂ MoSb	3.78	1.08	<i>ti</i>	<i>cr</i>	0.08	1.34	0.08	-0.25	0.52	-0.336	0.010	-0.95	11
Fe ₂ PdSb	3.86	1.06	<i>tr</i>	<i>ti</i>	0.10	5.57	-0.21	-0.49	0.40	-0.213	0.172	-1.01	1268
Fe ₂ PtIn	3.97	0.96	<i>ti</i>	<i>ci</i>	0.07	5.24	-0.39	-0.50	-0.32	-1.160	0.154	-3.50	1220
Fe ₂ PtGe	3.68	1.10	<i>tr</i>	<i>ti</i>	0.10	5.25	-0.26	-0.46	0.26	1.487	0.170	3.85	1081
Fe ₂ PtSb	3.87	1.06	<i>tr</i>	<i>ti</i>	0.17	5.61	-0.28	-0.56	0.43	1.036	0.173	2.25	1317
Co ₂ NiAl	3.67	0.92	<i>ti</i>	<i>tr</i>	0.13	2.65	-0.52	-0.44	-0.66	-0.629	0.052	-2.40	756
Co ₂ NiGa	3.69	0.92	<i>ti</i>	<i>tr</i>	0.10	2.73	-0.57	-0.36	-0.74	-0.618	0.054	-2.33	721
Co ₂ CuGa	3.70	0.94	<i>ti</i>	<i>cr</i>	0.05	2.58	-0.65	-0.64	-0.72	-0.087	0.047	-0.45	697
Co ₂ CoSi	3.58	0.93	<i>t</i>	<i>h</i>	0.07	2.79	-0.66	-0.60	-0.75	-0.599	0.062	-2.48	622
Co ₂ NiSi	3.55	0.96	<i>ti</i>	<i>tr</i>	0.05	2.23	-0.51	-0.42	-0.65	0.347	0.039	1.15	589
Co ₂ CoGe	3.64	0.94	<i>t</i>	<i>h</i>	0.10	2.99	-0.72	-0.68	-0.78	-0.634	0.066	-2.48	616
Co ₂ CoSn	3.81	0.94	<i>t</i>	<i>c</i>	0.08	3.17	-0.76	-0.74	-0.81	-0.640	0.065	-2.17	603
Co ₂ NiSn	3.80	0.95	<i>ti</i>	<i>tr</i>	0.09	2.49	-0.55	-0.48	-0.68	-0.066	0.040	-0.33	554
Co ₂ FeSb	3.97	0.85	<i>tr</i>	<i>cr</i>	0.14	4.81	-0.18	0.02	-0.49	-0.369	0.147	-1.55	849
Co ₂ CoSb	3.77	0.95	<i>t</i>	<i>h</i>	0.07	2.49	-0.51	-0.41	-0.67	0.129	0.041	0.28	455
Co ₂ MoGa	3.80	0.92	<i>tr</i>	<i>cr</i>	0.12	0.79	-0.11	-0.10	-0.13	0.053	0.004	0.15	6
Co ₂ PdGa	3.82	0.92	<i>ti</i>	<i>tr</i>	0.18	2.78	-0.50	-0.35	-0.66	-0.843	0.051	-2.79	785
Co ₂ RhIn	3.89	0.96	<i>ti</i>	<i>ci</i>	0.10	3.47	-0.78	-0.74	-0.86	-0.653	0.072	-2.05	729
Co ₂ PdIn	3.93	0.95	<i>ti</i>	<i>tr</i>	0.16	2.96	-0.74	-0.65	-0.84	-0.923	0.051	-2.71	758
Co ₂ RhGe	3.78	0.92	<i>ti</i>	<i>ci</i>	0.23	2.69	-0.67	-0.53	-0.81	-0.734	0.049	-2.52	624
Co ₂ PdGe	3.76	0.96	<i>ti</i>	<i>tr</i>	0.17	2.49	-0.59	-0.47	-0.72	0.230	0.041	0.60	653
Co ₂ MoSn	4.07	0.84	<i>tr</i>	<i>cr</i>	0.07	2.12	0.62	0.45	0.78	0.009	0.027	-0.05	97
Co ₂ RhSn	3.92	0.93	<i>ti</i>	<i>ci</i>	0.23	2.87	-0.72	-0.66	-0.81	-0.654	0.049	-2.01	664
Co ₂ PdSn	3.89	0.97	<i>ti</i>	<i>tr</i>	0.15	2.54	-0.55	-0.50	-0.65	-0.025	0.038	-0.18	610
Co ₂ RuSb	3.87	0.95	<i>ti</i>	<i>ci</i>	0.17	2.43	-0.37	-0.24	-0.57	0.192	0.036	0.45	446
Co ₂ RhSb	3.88	0.95	<i>ti</i>	<i>tr</i>	0.25	2.34	-0.62	-0.53	-0.76	0.403	0.033	1.07	536
Co ₂ WGa	3.88	0.87	<i>tr</i>	<i>cr</i>	0.13	1.07	0.49	0.48	0.51	0.812	0.008	2.54	64
Co ₂ OsGa	3.70	0.97	<i>tr</i>	<i>ti</i>	0.06	0.57	-0.49	-0.47	-0.50	-0.620	0.002	-2.03	0
Co ₂ IrGa	3.77	0.95	<i>ti</i>	<i>ci</i>	0.05	3.09	-0.63	-0.56	-0.73	-3.306	0.063	-10.61	557
Co ₂ PtGa	3.80	0.94	<i>ti</i>	<i>tr</i>	0.18	2.82	-0.69	-0.68	-0.75	-3.745	0.052	-11.79	748
Co ₂ IrIn	3.82	1.02	<i>ti</i>	<i>tr</i>	0.11	3.37	-0.76	-0.71	-0.84	-2.828	0.067	-8.16	581
Co ₂ PtIn	3.90	0.98	<i>ti</i>	<i>tr</i>	0.14	3.01	-0.80	-0.78	-0.86	-2.794	0.053	-7.84	715
Co ₂ IrGe	3.76	0.95	<i>ti</i>	<i>ci</i>	0.24	2.51	-0.54	-0.41	-0.69	-2.731	0.042	-8.80	528
Co ₂ PtGe	3.76	0.97	<i>ti</i>	<i>tr</i>	0.16	2.44	-0.58	-0.53	-0.68	-0.404	0.039	-1.38	648
Co ₂ WSn	4.07	0.84	<i>tr</i>	<i>cr</i>	0.11	1.76	0.14	-0.11	0.41	-4.368	0.018	-12.41	43
Co ₂ IrSn	3.88	0.97	<i>ti</i>	<i>tr</i>	0.20	2.79	-0.66	-0.59	-0.76	-2.963	0.046	-8.51	596
Co ₂ PtSn	3.89	0.98	<i>ti</i>	<i>tr</i>	0.11	2.52	-0.55	-0.52	-0.66	0.206	0.037	0.47	624
Co ₂ WSb	3.76	1.07	<i>ti</i>	<i>tr</i>	0.05	1.12	0.52	0.43	0.67	-0.664	0.007	-1.89	5
Co ₂ OsSb	3.86	0.97	<i>ti</i>	<i>ci</i>	0.23	2.19	-0.22	-0.17	-0.34	-2.806	0.029	-8.14	428
Co ₂ IrSb	3.86	0.97	<i>ti</i>	<i>tr</i>	0.27	2.34	-0.43	-0.37	-0.55	-0.801	0.033	-2.40	550
Pd ₂ FeGa	3.96	0.93	<i>tr</i>	<i>cr</i>	0.09	3.22	-0.35	-0.14	-0.60	0.041	0.061	-0.06	457
Pd ₂ CoGa	3.92	0.93	<i>tr</i>	<i>cr</i>	0.21	1.94	-0.50	-0.35	-0.67	-0.383	0.022	-1.16	341
Pd ₂ FeIn	4.09	0.93	<i>tr</i>	<i>cr</i>	0.06	3.20	-0.39	-0.28	-0.62	0.257	0.054	0.51	543
Pd ₂ CoIn	4.03	0.95	<i>tr</i>	<i>cr</i>	0.21	1.94	-0.39	-0.19	-0.62	-0.483	0.020	-1.30	261
Pd ₂ FeGe	3.91	0.97	<i>tr</i>	<i>cr</i>	0.10	3.12	-0.70	-0.26	-0.86	0.150	0.057	0.26	245
Pd ₂ CoGe	3.90	0.95	<i>tr</i>	<i>ti</i>	0.11	1.71	-0.67	-0.46	-0.80	1.389	0.017	3.90	259
Pd ₂ CoSn	4.03	0.95	<i>tr</i>	<i>ti</i>	0.10	1.61	-0.71	-0.46	-0.84	1.064	0.014	2.70	217
Pd ₂ CoSb	4.12	0.89	<i>ti</i>	<i>ci</i>	0.05	1.25	-0.44	-0.56	-0.18	-0.060	0.008	-0.18	0
Rh ₂ FeGe	4.01	0.85	<i>tr</i>	<i>cr</i>	0.21	3.83	-0.49	-0.09	-0.78	-0.898	0.090	-2.89	516
Rh ₂ CoGe	3.95	0.87	<i>tr</i>	<i>cr</i>	0.19	2.22	-0.57	-0.13	-0.80	-0.754	0.031	-2.34	487
Rh ₂ FeSn	4.16	0.85	<i>tr</i>	<i>cr</i>	0.19	3.89	-0.61	-0.29	-0.83	-0.985	0.083	-2.80	503
Rh ₂ CoSn	4.07	0.89	<i>tr</i>	<i>cr</i>	0.21	2.30	-0.61	-0.22	-0.84	-0.621	0.030	-1.75	511
Rh ₂ MnSb	4.15	0.87	<i>tr</i>	<i>cr</i>	0.20	4.17	0.00	0.07	-0.04	-0.469	0.094	-1.46	336
Rh ₂ FeSb	4.05	0.91	<i>tr</i>	<i>cr</i>	0.37	3.38	-0.42	0.08	-0.76	-0.164	0.064	-0.60	472
Rh ₂ CoSb	4.01	0.92	<i>tr</i>	<i>ti</i>	0.44	2.05	-0.22	0.23	-0.59	0.685	0.024	1.79	48

TABLE I. (Continued.)

	a (Å)	c'	s_1	s_2	E_{21} (eV)	m (μ_B)	SP_t	SP_1	SP_2	K_{mc} $\frac{\text{meV}}{\text{f.u.}}$	K_{sh} $\frac{\text{meV}}{\text{f.u.}}$	K_v $\frac{\text{MJ}}{\text{m}^3}$	T_C (K)
Ru ₂ CoGa	3.72	1.02	<i>ti</i>	<i>cr</i>	0.11	1.06	-0.48	-0.49	-0.50	-0.064	0.007	-0.22	34
Ru ₂ CoIn	3.89	0.99	<i>ti</i>	<i>cr</i>	0.22	1.28	-0.46	-0.50	-0.43	-0.009	0.009	-0.05	80
Ru ₂ CoSn	3.90	0.98	<i>ti</i>	<i>cr</i>	0.07	0.04	-0.17	-0.19	-0.13	0.000	0.000	0.00	0
Ni ₂ FeGa	3.67	0.96	<i>tr</i>	<i>cr</i>	0.07	3.30	-0.31	-0.30	-0.44	0.084	0.077	0.02	524
Ni ₂ CoSi	3.60	0.92	<i>ti</i>	<i>ci</i>	0.11	0.89	-0.67	-0.69	-0.68	0.156	0.006	0.56	105
Ni ₂ CoGe	3.66	0.93	<i>ti</i>	<i>tr</i>	0.11	1.03	-0.68	-0.70	-0.68	0.124	0.008	0.41	94

one spin channel, σ_1 , and does not have such states in another spin channel, σ_2 . [The Γ -Z line in BZ is the line along the k_z direction with in-plane wave vector $\mathbf{k}_{\parallel} = (k_x, k_y) = 0$.] It is well known [24,25] that the smallest (at given \mathbf{k}_{\parallel}) attenuation constant $\gamma(\mathbf{k}_{\parallel})$ of MgO for evanescent states propagating along the z direction with energies within the MgO band gap reaches a minimum at $\mathbf{k}_{\parallel} = 0$. When $|\mathbf{k}_{\parallel}|$ increases, $\gamma(\mathbf{k}_{\parallel})$ increases as $\gamma(\mathbf{k}_{\parallel}) = \gamma_0 + \alpha k_{\parallel}^2$ (with $\alpha > 0$). Therefore, at $E = E_F$, the evanescent states of ME in σ_1 spin channel that propagate along the z direction with $\mathbf{k}_{\parallel} = 0$ will decay inside MgO as $e^{-\gamma_0 z}$, while evanescent states in the σ_2 spin channel will decay as $e^{-(\gamma_0 + \alpha k_{\parallel}^2)z}$, with $|\mathbf{k}_{\parallel}| > 0$ since the σ_2 channel does not have states at E_F with $\mathbf{k}_{\parallel} = 0$. As a result the TMR increases exponentially with increasing MgO thickness, d_{MgO} : $\text{TMR} \propto \exp(2\alpha \tilde{\mathbf{k}}_{\parallel}^2 d_{\text{MgO}})$, where $\tilde{\mathbf{k}}_{\parallel}$ is the shortest vector \mathbf{k}_{\parallel} for which ME has states in the σ_2 spin channel at E_F . Such dependence on d_{MgO} is much stronger than the $\text{TMR} \propto d_{\text{MgO}}^n$ dependence arising from the symmetry filtering effect [24,25], where the power factor n can only take three values $n = 0, 1, 2$ for MTJ systems with square symmetry in the xy plane [26].

Table II shows the values of $E_z = E_c - E_F$ for 26 compounds that have a band gap along the Γ -Z line ($|E_z| > 0$) in only one spin channel, and for Fe₂CuAl and Fe₂CuGa that have a band gap ($|E_z| > 0$) in both spin channels. Here E_c is the closest to E_F energy among energies of states in corresponding spin channel with \mathbf{k} along the Γ -Z line. The value of $|E_z|$ characterizes the strength of the BZ filtering effect since larger $|E_z|$ leads to larger value of $\tilde{\mathbf{k}}_{\parallel}$ and, therefore, to faster TMR increase with d_{MgO} . Also, large values of $|E_z|$ make the existence of the BZ filtering effect less susceptible to the details of calculations (choice of the DFT functional, variations of the lattice constants, etc.) as well as to the experimental conditions (effects of disorder, finite temperature, finite applied bias, etc.).

The majority (minority) spin channel with $|E_z| > 0$ is indicated in Table II by an up (down) arrow. Positive (negative) E_z indicates that the band closest to E_F is located above (below) E_F . For each compound E_z was calculated by three different methods: the pseudopotential PAW approach implemented in the VASP program with the PBE GGA/DFT functional, the full-potential all-electron LMTO approach [27] with Barth-Hedin LDA/DFT functional [28], and the quasiparticle self-consistent GW (QSGW) method that is known to describe band gaps and other properties of materials with moderate e - e correlations significantly better than DFT [29–31]. Three values of E_z are indicated as E_z^{GGA} , E_z^{LDA} , and E_z^{GW} , correspondingly, in Table II. The majority and minority bands along the Γ -Z line calculated by the GGA/DFT,

LDA/DFT, and QSGW methods are shown for 28 compounds in Figs. 2–5 by red, green, and blue, correspondingly.

Table II shows that for Mn₂CuSn, Fe₂CuGa, and Fe₂MoSb compounds $|E_z^{\text{GGA}}| > 0$ and $|E_z^{\text{LDA}}| > 0$, while the gap along Γ -Z closes in QSGW, $|E_z^{\text{GW}}| = 0$. On the other hand, for Ni₂CoGe, Co₂NiGa, and Co₂PdGa compounds $|E_z^{\text{GGA}}| = |E_z^{\text{LDA}}| = 0$, while the gap along the Γ -Z line opens in QSGW, $|E_z^{\text{GW}}| > 0$. Thus, beyond DFT calculations are necessary in order to accurately estimate E_z . Note that for one of the most studied tetragonal Heusler compounds, Mn₃Ge, the QSGW value $|E_z^{\text{GW}}| = 0.03$ eV is significantly smaller than the GGA

TABLE II. The values of E_z calculated by GGA, LDA, and QSGW methods are shown for 28 tetragonal Heusler compounds.

		E_z^{GGA} (eV)	E_z^{LDA} (eV)	E_z^{GW} (eV)
Mn ₃ Ge	↓	-0.08	-0.05	-0.03
Mn ₃ Sn	↓	-0.22	-0.17	0.20
Mn ₃ Sb	↓	-0.10	-0.12	0.29
Mn ₂ CoSn	↓	-0.05	-0.03	-0.02
Mn ₂ NiSi	↑	0.16	0.06	-0.14
Mn ₂ NiGe	↑	-0.23	0.11	0.09
Mn ₂ CuSi	↓	0.41	0.33	0.41
Mn ₂ CuGe	↓	0.42	0.49	0.17
Mn ₂ CuSn	↓	0.08	0.15	0.00
Mn ₂ MoSn	↑	-0.16	-0.16	-0.27
Mn ₂ MoSb	↓	-0.44	-0.40	-0.41
Mn ₂ WSb	↓	-0.58	-0.49	-0.56
Mn ₂ OsGe	↓	-0.05	-0.03	0.16
Ni ₂ CoGe	↓	0.00	0.00	0.08
Fe ₂ MnSn	↓	0.24	0.08	0.23
Fe ₂ NiSi	↓	0.27	0.18	0.37
Fe ₂ NiGe	↓	0.39	0.28	-0.29
Fe ₂ NiSn	↓	-0.30	-0.28	-0.34
Fe ₂ NiSb	↓	0.11	0.06	0.21
Fe ₂ CuAl	↑	0.06	0.17	0.11
Fe ₂ CuAl	↓	-0.22	-0.31	-0.12
Fe ₂ CuGa	↑	-0.09	0.03	0.00
Fe ₂ CuGa	↓	-0.06	-0.14	0.00
Fe ₂ MoSb	↓	0.14	0.12	0.00
Fe ₂ PdGe	↓	-0.36	-0.41	-0.28
Fe ₂ PdSb	↓	0.18	0.14	0.26
Fe ₂ PtGe	↓	-0.31	-0.34	-0.16
Fe ₂ PtSb	↓	0.09	0.09	0.16
Co ₂ NiGa	↓	0.00	0.00	-0.05
Co ₂ PdGa	↓	0.00	0.00	-0.03

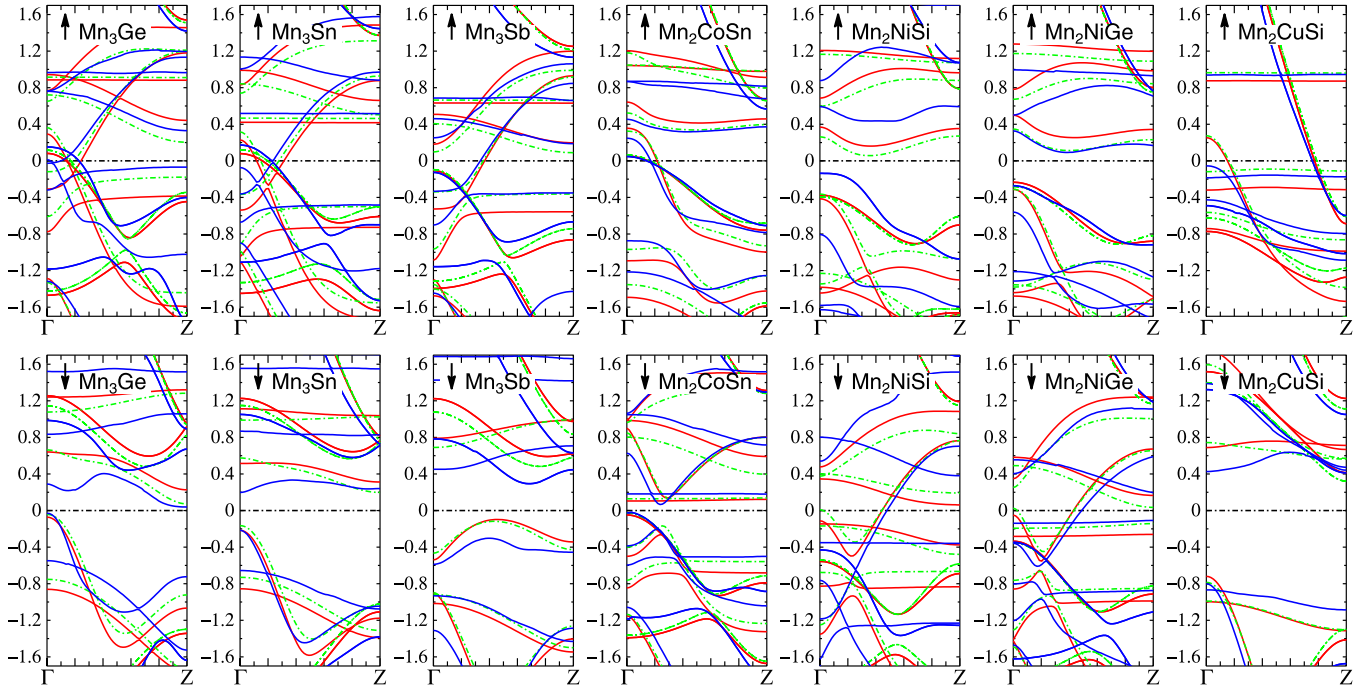


FIG. 2. Majority (\uparrow) and minority (\downarrow) bands of Mn_3Ge , Mn_3Sn , Mn_3Sb , Mn_2CoSn , Mn_2NiSi , Mn_2NiGe , and Mn_2CuSi along the Γ -Z line. GGA, LDA, and QSGW bands are shown by the red, green, and blue. Vertical scale is $E - E_F$ (eV).

value $|E_z^{\text{GGA}}| = 0.08$ eV. Therefore, high TMR values predicted previously by DFT calculations in $\text{Mn}_3\text{Ge}/\text{MgO}/\text{Mn}_3\text{Ge}$ MTJ [9,12,32] are expected to be significantly lower if calculated by more accurate beyond DFT methods.

Table II shows that there are 17 compounds with relatively large $|E_z^{\text{GW}}| > 0.15$ eV. These compounds are expected to exhibit large TMR in MTJ devices. Among these 17 compounds

13 have PMA ($K_v > 0$) and 10 have strong PMA with $K_v > 0.5$ MJ/m³: Mn_3Sn , Mn_3Sb , Mn_2CuSi , Mn_2CuGe , Mn_2OsGe , Fe_2MnSn , Fe_2NiSn , Fe_2PdGe , Fe_2PtGe , and Fe_2PtSb . These ten compounds constitute our “best” candidates for STT-MRAM applications identified in the present paper. Note, that five Fe-based compounds in this list have high $T_C > 1000$ K (see Table I).

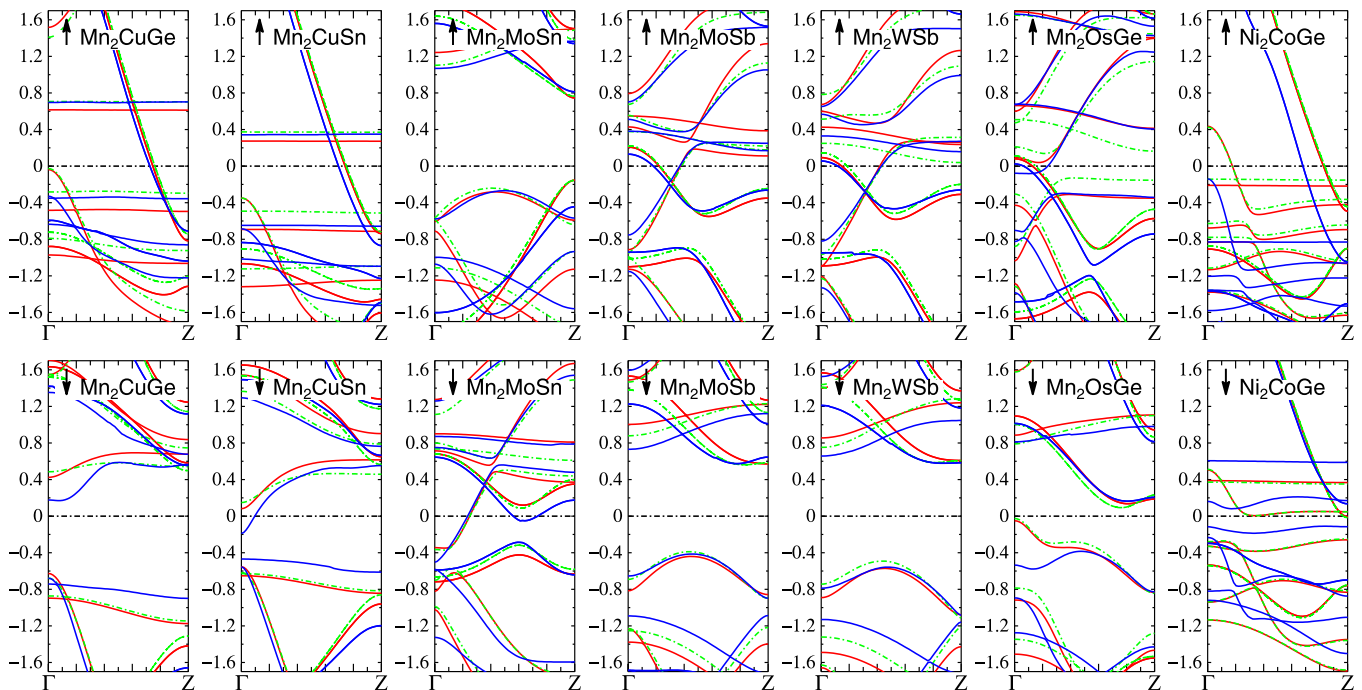


FIG. 3. Majority (\uparrow) and minority (\downarrow) bands of Mn_2CuGe , Mn_2CuSn , Mn_2MoSn , Mn_2MoSb , Mn_2WSb , Mn_2OsGe , and Ni_2CoGe along the Γ -Z line. GGA, LDA, and QSGW bands are shown by red, green, and blue. Vertical scale is $E - E_F$ (eV).

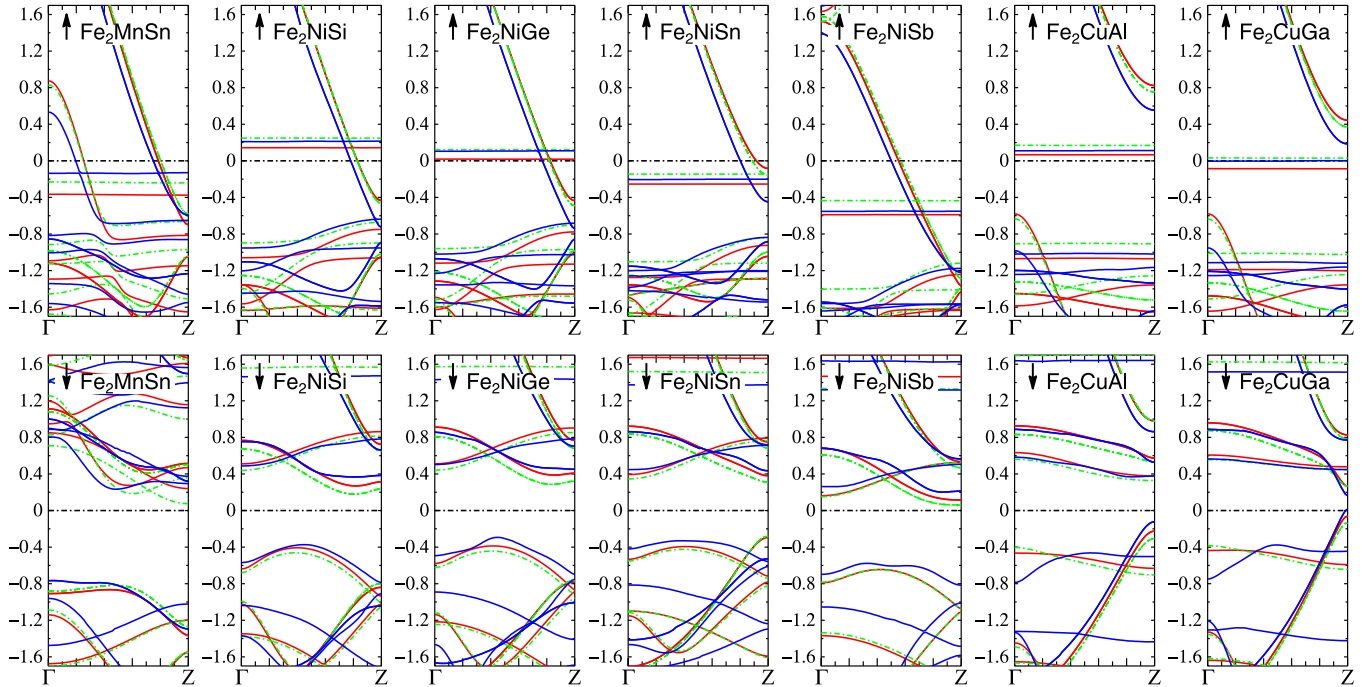


FIG. 4. Majority (\uparrow) and minority (\downarrow) bands of Fe_2MnSn , Fe_2NiSi , Fe_2NiGe , Fe_2NiSn , Fe_2NiSb , Fe_2CuAl , and Fe_2CuGa along the Γ -Z line. GGA, LDA, and QSGW bands are shown by red, green, and blue. Vertical scale is $E - E_F$ (eV).

D. Compounds with expected large TMR due to large spin polarization

Large spin polarization of bulk Heusler compounds also could result in enhanced TMR values. We identified 11 compounds— Mn_3In , Mn_2FeIn , Mn_3Ge , Mn_2CoGe , Mn_3Sn , Mn_2CoSn , Mn_2RuSn , Mn_2IrIn , Mn_2OsGe , Mn_2OsSn , and Pd_2CoS —in Table I that have large total spin polarization,

$|SP_I| > 0.7$ and, simultaneously, PMA with $K_v > 0.6 \text{ MJ/m}^3$. These 11 compounds constitute our “second best” list of candidates for STT-MRAM applications. Eight compounds from this list have very strong PMA with $K_v \geq 1.8 \text{ MJ/m}^3$ and nine compounds have $T_C > 460 \text{ K}$. Note that the sign of the spin polarization is the same for both termination layers for these 11 compounds (see Table I) which is important since, as

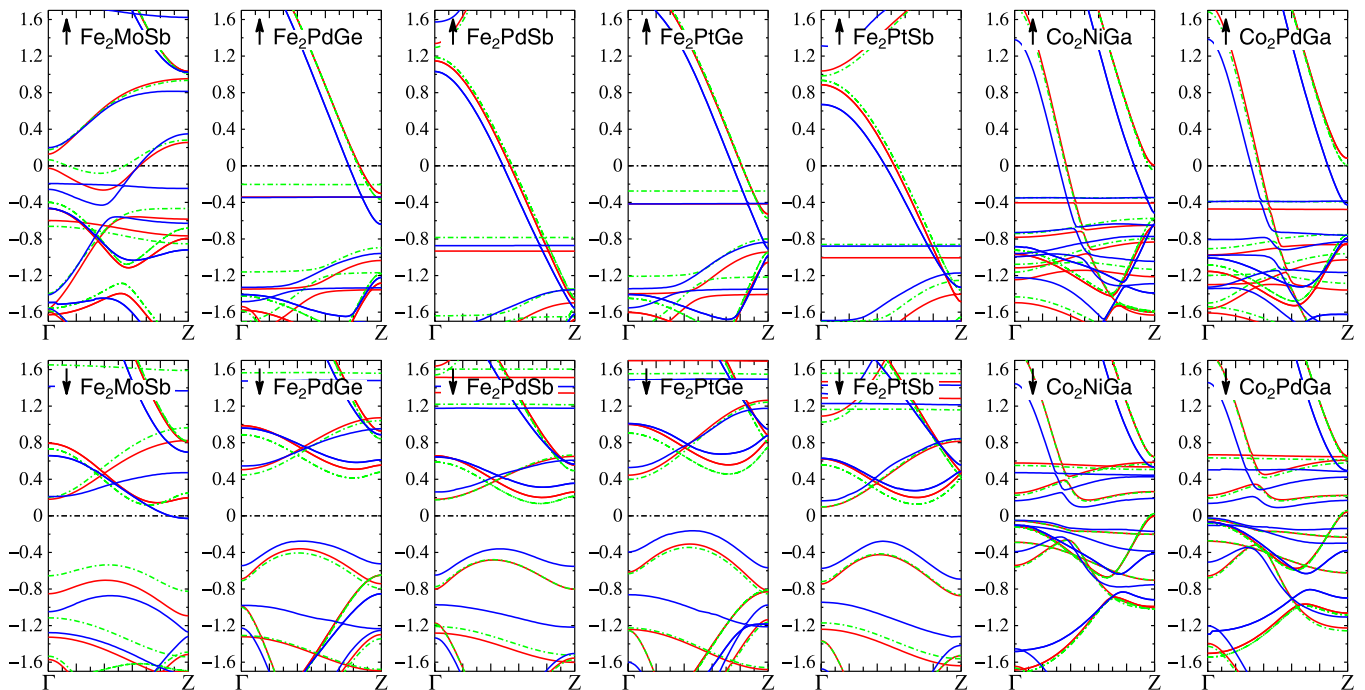


FIG. 5. Majority (\uparrow) and minority (\downarrow) bands of Fe_2MoSb , Fe_2PdGe , Fe_2PdSb , Fe_2PtGe , Fe_2PtSb , Co_2NiGa , and Co_2PdGa along the Γ -Z line. GGA, LDA, and QSGW bands are shown by red, green, and blue. Vertical scale is $E - E_F$ (eV).

was discussed in Ref. [12], in real devices both terminations can be randomly realized at the ME/MgO interface. If the signs of SP_1 and SP_2 were different, the spin polarization of the tunneling current in areas with different terminations would have different sign, thereby reducing the total TMR.

Two Heusler compounds— Mn_3Sn and Mn_2OsGe —belong to both lists since they simultaneously have large spin polarization and BZ filtering conditions. Unfortunately, both effects tend to cancel each other for these compounds. For small d_{MgO} (when the effect of BZ filtering is small) the tunneling (inside MgO barrier) spin polarization for these compounds is expected to be dominated by the spin polarization of bulk Heuslers and have negative sign, while for large d_{MgO} the sign of the tunneling spin polarization should switch to positive due to the increasing role of the BZ filtering effect.

We note that for ideal junctions the candidate materials from the best list should have higher priority for experimental study as compared to the materials from the second best list since TMR should grow exponentially with d_{MgO} for MTJs with electrodes from the best list due to the BZ filtering effect, while the spin polarization induced enhancement of the TMR for MTJs with electrodes from the second best list is not expected to vary much when d_{MgO} increases. On the other hand, the BZ filtering effect relies on the existence of a well-defined surface Brillouin zone and in real devices (with lattice mismatch and disorder) the TMR enhancement due to the BZ filtering effect could be suppressed. The spin polarization induced enhancement of the TMR is less sensitive to the disorder and lattice mismatch. Therefore, both the best list and the second best list of candidate materials should be experimentally explored since the winner material could belong to any of these lists.

III. EXPERIMENTAL IDENTIFICATION OF TETRAGONAL HEUSLER COMPOUNDS WITH PERPENDICULAR MAGNETIC ANISOTROPY

We performed experimental measurements for 32 Heusler compounds. 20–30-nm-thick films of these Heusler compounds were prepared by either dc-magnetron sputtering or ion-beam deposition in an ultrahigh-vacuum chamber (base pressure 4×10^{-10} Torr). Various buffer layers of Si/SiO₂/TaN/IrM₃, Si/SiO₂/TaN/IrM₃/TaN, MgO(001), MgO(001)/Cr, MgO(001)/Cr/Ir, and MgO(001)/Cr/IrMn₃ were used to reduce lattice mismatch between Heusler compounds and the substrate. Each Heusler compound was grown at several different substrate temperatures, typically 100 °C–600 °C, and magnetic properties of them were measured by a Quantum Design superconducting quantum interference device vibrating sample magnetometer in magnetic fields of up to ± 7 T. More details on the experimental setup can be found in Ref. [12].

A comparison of the experimental results with theoretical predictions for 17 compounds is presented in Table III. Experimental measurements confirm stable tetragonal phase for ten compounds that are predicted to be tetragonal, and stable cubic phase for six compounds that are predicted to be cubic. Fe₂MnGa was found to be tetragonal in experiment, while predicted to be “unstable” cubic in theory (the tetragonal phase of Fe₂MnGa is only 0.03 eV higher in energy than the

TABLE III. The experimental ($c^{\text{expt.}}$, $m^{\text{expt.}}$, $K_v^{\text{expt.}}$) and calculated ($c^{\text{theor.}}$, $m^{\text{theor.}}$, $K_v^{\text{theor.}}$) values of the lattice constant, c , magnetic moment, m , and anisotropy constant, K_v , for 17 measured Heusler compounds. $s^{\text{expt.}}$ and $s^{\text{theor.}}$ label experimentally found and the lowest-energy DFT-calculated structures (except for Fe₂MnGa where DFT results are shown for a tetragonal phase that is 0.03 eV higher in energy than the cubic one). The symbol “>” in some of the experimental $K_v^{\text{expt.}}$ means that the shown value is a lower bound for $K_v^{\text{expt.}}$.

	$s^{\text{expt.}}$	$c^{\text{expt.}}$ (Å)	$m^{\text{expt.}}$ emu cm ³	$K_v^{\text{expt.}}$ MJ m ³	$s^{\text{theor.}}$	$c^{\text{theor.}}$ (Å)	$m^{\text{theor.}}$ emu cm ³	$K_v^{\text{theor.}}$ MJ m ³
Mn ₃ Ge	tet	7.2	130	>0.45	tet	7.09	190	2.48
Mn ₃ Sn	tet	7.6	150	>0.52	tet	7.47	167	2.65
Mn ₂ FeGa	tet	7.2	300	>1.05	tet	7.23	158	1.59
Mn ₂ CoSn	tet	6.4	350	0.26	tet	7.13	166	0.95
Mn ₂ CuSb	tet	6.5	120	0.06	tet	7.80	870	2.18
Mn ₂ RuSn	tet	7.3	175	>0.00	tet	7.70	0	1.14
Mn ₂ WSb	tet		250	>0.00	tet	8.29	9	−2.34
Fe ₂ MnGa	tet	7.0	650	0.32	tet	6.51	1335	0.30
Ni ₂ CoGe	tet	7.1	150	>0.00	tet	6.81	209	0.41
Rh ₂ FeSb	tet	7.0	300	>0.15	tet	7.37	518	−0.60
Rh ₂ CoSb	tet	7.1	200	>0.70	tet	7.38	320	1.79
Mn ₃ Si	cub	5.7	0		cub	5.64	206	
Co ₂ MnAl	cub	5.7			cub	5.70	803	
Co ₂ MnGe	cub	5.8	1000		cub	5.74	979	
Co ₂ MnSi	cub	5.6			cub	5.63	1040	
Co ₂ FeAl	cub	5.6			cub	5.70	997	
Ni ₂ MnGe	cub	5.8	660		cub	5.81	742	

cubic phase [15]). Note that 9 out of 11 tetragonal compounds in Table III have PMA in both the experiment and theory (exceptions are Mn₂WSb and Rh₂CoSb where $K_v^{\text{theor.}} < 0$, but $K_v^{\text{expt.}} > 0$).

The remaining 15 out of 32 measured compounds include Co₂NiGe, Co₂RhSb, Mn₂NiSb, Mn₂CuSn, Fe₂CuAl, Co₃Sn, Co₃Sb, Co₃Ge, Fe₂CuSn, Mn₂CuSi, Co₂IrSb, Ru₂CoGa, Mn₂PtSb, Fe₂PtSb, and Mn₂OsSn. All these compounds were predicted to have tetragonal phase by DFT calculations but were found to be cubic in experiment. We attribute this discrepancy to the effects of disorder that favor high-symmetry cubic phase. Since DFT calculations assume zero temperature, the finite temperature effects could also contribute to this discrepancy.

Despite disagreement between theoretical predictions and experimental results for some studied compounds, experimental confirmation of tetragonality for 10 out of 26 measured compounds that were predicted to be tetragonal by the theory shows that DFT calculations for ideal systems (without taking into account disorder and finite temperature effects) can still correctly predict tetragonality in a significant share of studied cases. Moreover, for the majority of found tetragonal compounds (9 out of 11) theory also correctly predicted PMA.

IV. CONCLUSION

In conclusion, we performed DFT calculations for 286 Heuslers in cubic, tetragonal, and hexagonal phases, and

identified 116 stable tetragonal compounds. Out of these 116 materials we identified 19 potential candidates for electrodes for STT-MRAM MTJ devices. These 19 compounds simultaneously have PMA (with high $K_v > 0.9$ MJ/m³ for 15 of these materials) and expected to have enhanced TMR either due to the strong BZ filtering effect ($|E_z| > 0.15$ eV) or due to the high spin polarization ($|SP_z| > 0.7$). The QSGW calculations of the band structure performed for 28 stable tetragonal compounds that satisfy the BZ filtering conditions show that beyond DFT methods are needed to accurately evaluate the strength (and even existence) of the BZ filtering effect.

We performed experimental measurements for 32 Heusler compounds. To the best of our knowledge, the majority of the tetragonal compounds presented in Table III are experimentally identified as tetragonal compounds with PMA for the first time (exceptions are known tetragonal compounds Mn₃Ge [7], Mn₂FeGa, and Fe₂MnGa [33]). Our experimental results show

that DFT calculations can correctly predict both tetragonality and PMA in a significant share of studied cases. Therefore, one can expect that experimental measurements for Heusler compounds theoretically predicted to be tetragonal with PMA in the present work (as well as in further theoretical studies) will result in experimental identification of a significant number of stable tetragonal Heusler compounds with PMA suitable for spintronics applications.

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