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Density functional theory investigation on lattice dynamics, elastic properties and origin of vanished magnetism in Heusler compounds CoMnVZ (Z = Al, Ga)*

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The lattice dynamics, elastic properties and the origin of vanished magnetism in equiatomic quaternary Heusler compounds CoMnVZ (Z = Al, Ga) are investigated by first principle calculations in this work. Due to the similar constituent atoms in CoMnVAl and CoMnVGa compounds, they are both stable in LiMgPdSn-type structure with comparable lattice size, phonon dispersions and electronic structures. Comparatively, we find that CoMnVAl is more structurally stable than CoMnVGa. Meanwhile, the increased covalent bonding component in CoMnVAl enhances its mechanical strength and Vickers hardness, which leads to better comprehensive mechanical properties than those of CoMnVGa. Practically and importantly, structural and chemical compatibilities at the interface make non-magnetic semiconductor CoMnVAl and magnetic topological semimetals Co_2MnAl/Ga more suitable to be grown in heterostructures. Owing to atomic preferential occupation in CoMnVAl/Ga, the localized atoms Mn occupy C (0.5, 0.5, 0.5) Wyckoff site rather than B (0.25, 0.25, 0.25) and D (0.75, 0.75, 0.75) Wyckoff sites in LiMgPdSn-type structure, which results in symmetric band filling and consequently drives them to be non-magnetic. Correspondingly, by tuning localized atoms Mn to occupy B (0.25, 0.25, 0.25) or/and D (0.75, 0.75, 0.75) Wyckoff sites in off-stoichiometric Co–Mn–V–Al/Ga compounds and keeping the total valence electrons as 24, newly compensated ferrimagnetic compounds are theoretically achieved. We hope that our work will provide more choices for spintronic applications.

Keywords: Heusler compounds CoMnVAl/Ga, first principles calculations, lattice dynamics, elastic properties, nonmagnetic semiconductor

PACS: 31.15.A–, 75.20.Hr, 62.20.–x, 63.20.dk **DOI:** 10.1088/1674-1056/ac0a6a

1. Introduction

Heusler compounds have been being the research focus of multiple functional materials with novel properties in these years. Their outstanding properties range from ferromagnetic shape memory effect in conventional[1] and all-d-metal[2,3] Heusler compounds, spin gapless semiconductor,[4] half metallicity[5] and so on. Especially in recent years, the discovery of magnetic topological semimetals in half metallic Heusler compounds further extends their research field and makes them one of the most attractive fields of research in condensed matter physics and materials science.[6–11] For magnetic topological semimetals, their symmetries and electronic structures depend sensitively on their magnetic structures and magnetization directions. Thus, it is convenient and realistic to manipulate their topological properties and phase transition by applying an external magnetic field, which makes Heusler half metallic compounds highly useful in the design of spintronic devices.[11] Practically, these kinds of materials are usually incorporated in spintronic devices in the form of films or multilayers. At the interface between these materials and semiconductors, the d–d hybridization creating the half-metal gap and determining magnetism stops abruptly and consequently results in d-like dangling bond states inside the gap. Thus, the presence of interface states in sandwiching structure is by construction prone to reduce or even invert the spin polarization and impact on their magnetism at the interface.[12] Therefore, interface state between these materials and semiconductors is an important issue for technical application. In order to obtain high quality heterostructures for spintronic devices, the sandwiching structures with suitable semiconductors are necessary. For Heusler compounds, they possess extremely wide range of electronic properties with rather similar geometric structure. It provides the possibility to construct the heterostructures where magnetic functional compounds and semiconductors are both Heusler building blocks.[13] By this, both topological and half-metallic properties can be smoothly preserved through the interface. Thus, to investigate Heusler-type nonmagnetic semiconductor will be of importance from both fundamental re-

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search and application points of view.

Currently, $L2_1$-type half-metallic Heusler compounds Co_2MnAl/Ga have been theoretically predicted as novel magnetic topological semimetals with bright future as spintronic candidate materials.[7–10] Doping one Co atom by V atom in primitive cell of Co_2MnAl/Ga, the LiMgPdSn-type CoMnVAl/Ga can be obtained with just 24 valence electrons. According to the well-known Slater–Pauling rule,[14,15] CoMnVAl/Ga prefer to be nonmagnetic semiconductor.[13,16] Possibly, similar structures and chemical compatibilities between Co_2MnAl/Ga and CoMnVAl/Ga make them suitable heterostructures with smooth interface transition. The bonding character could be kept through the interface and consequently the properties of parent compound can also be preserved. Previous calculations verified that, among all possible stackings in the heterostructure composed by Co_2MnAl and CoMnVAl, Co–Co/V–Al and Co–Mn/Mn–Al were identified as energetically favorable interfaces, and suitable to construct heterostructures for spintronic devices.[13] To further experimentally realize these heterostructures, except the parent compounds, the structure stability, mechanical properties and electronic structure of non-magnetic CoMnVAl/Ga deserve detailed investigation. Additionally, in Heusler family, compounds with 24 valence electrons can be also the fully compensated ferrimagnetic half metals (FCFHMs).[17,18] Further deep understanding of the origin of the vanished magnetic moment in equiatomic quaternary CoMnVAl/Ga will be helpful to tune the magnetic exchange interactions and induce the formation of FCFHMs in off-stoichiometric Co–Mn–V–Al/Ga compounds. Hopefully, the same constituent atoms and similar crystal structures between non-magnetic CoMnVAl/Ga and off-stoichiometric Co–Mn–V–Al/Ga FCFHMs will promote the outstanding heterostructures for spin electronic devices. In this work, we will firstly investigate the crystal structure, electronic structure, lattice dynamics properties and mechanical properties of CoMnVAl/Ga, and then the origin of vanished magnetic moment in CoMnVAl/Ga. Based on the results, the possible scheme will be illustrated to get fully compensated ferrimagnetic compounds in off-stoichiometric Co–Mn–V–Al/Ga. One expects the current work can provide more choices or suggestions to explore outstanding candidate materials for spintronic applications.

The paper is organized as follows: Section 2 contains the first-principle calculation methods and details. Section 3 presents the calculated results, including: (1) the crystal structure at ground state; (2) calculated electronic structure; (3) lattice dynamics properties; and (4) the elastic properties. Section 4 presents a detailed discussion about the mechanical properties and the origin of vanished magnetism followed by theoretical design and realization of possible fully compensated ferrimagnetic compounds. Finally, the paper is summarized in Section 5.

2. Calculation methods and details

The possibilities offered by *ab initio* techniques are explored to investigate the lattice dynamics, elastic properties and the origin of vanished magnetism in Heusler compounds CoMnVZ (Z = Al, Ga). The computer simulations were carried out by the alloy theory formulated within the coherent potential approximation (CPA) as implemented in the exact muffin-tin orbitals (EMTO) method.[19–21] It is flexible enough to deal with atomic random occupation in off-stoichiometric Heusler compounds. This approach has been used in numerous applications on metals and alloys.[22–25] The details for elastic constant calculations were as the same as those in Ref. [26]. With the help of CASTEP code, the electron localization function distribution was investigated by the pseudopotential method. The energy convergence criterion was set to be 1×10^{-6} eV/atom. The density plane-wave cutoff is 500 eV. The interactions between the atomic core and the valence electrons were described by the ultrasoft pseudopotential.[27] During all the calculations, the exchange-correlation energy of electrons was dealt by GGA-PBE.[28] A k-point mesh of $15 \times 15 \times 15$ was selected.

3. Calculation results

3.1. Crystal structure at ground state

Both CoMnVAl and CoMnVGa in this work crystallize in LiMgPdSn-type structure,[13,29] as shown in Fig. 1. In Heusler family, the general chemical formula of LiMgPdSn-type quaternary compounds is $XX'YZ$, in which X, X', and Y are transition metal atoms and Z is main group element. The valence electrons of X are more than those of X' atoms while the valence electrons of Y are less than those of both X and X'. Based on the atomic preferential occupation rule in Heusler compounds,[30] X, Y and X' atoms will occupy A, B, and C sites, respectively. Z atoms lie in the left D sites. Thus, in CoMnVAl/Ga compounds, Co and Mn atoms will occupy the A (0, 0, 0) and C (0.5, 0.5, 0.5) sites, respectively. While the V and Al/Ga atoms lie in the B (0.25, 0.25, 0.25) and D (0.75, 0.75, 0.75) sites, respectively. This atomic configuration has been confirmed by our theoretical calculation and other works.[13,16,29,31–33]

In Heusler compounds, there is strong p–d covalent hybridization between the first nearest neighboring (1nn) atomic pairs of transition metal atoms and main group elements.[34,35] Based on above atomic configuration, Co/Mn and Al/Ga are 1nn atomic pairs in LiMgPdSn-type CoMnVAl and CoMnVGa compounds. Thus, in this work the specific p–d covalent hybridization is between electrons in 3d orbital of Co/Mn atoms

and electrons in 3p orbital of Al or 4p orbital of Ga. This covalent hybridization contributes not only to structure but also functional properties, which will be discussed in this work.

Based on this atomic configuration, the first principle calculations have been carried out. The structural information at equilibrium state were obtained by geometry optimization. Based on them, the lattice constants along with results from experiments and other theory work, bulk modulus and its pressure derivative, and cohesive energy were achieved and listed in Table 1.

For cubic CoMnVAl and CoMnVGa compounds, their calculated lattice constants in this work are 5.75 Å and 5.77 Å, respectively. They are closed to other theoretic values calculated within generalized gradient approximation (GGA) of the exchange-correlation potential as parameterized by Perdew, Burke, and Ernzerhof (PBE).[33,36] Despite these lattice constants are underestimated when compared with experimental work, the theoretical and experimental lattice constants of CoMnVGa are always slightly bigger than those of CoMnVAl. These consistencies suggest current calculations are reasonable and it is safe to carry out the following discussions.

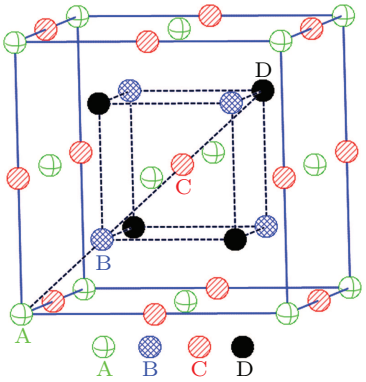


Fig. 1. The crystal structure of LiMgPdSn-type equiatomic quaternary Heusler compounds(space group $F\bar{4}3m$, 216) with four interpenetrating fcc sublattices of A, B, C, and D along the diagonal, whose coordinates are (0 0 0), (0.25 0.25 0.25), (0.5 0.5 0.5) and (0.75 0.75 0.75), respectively.

Table 1. Ground state properties, lattice constant A (in Å), bulk modulus B (in GPa), pressure derivative of the bulk modulus B', cohesive energies $E_{\rm coh}$ (in eV) for CoMnVAl and CoMnVGa compounds at theoretical equilibrium states.

Compounds		A	B	B'	$E_{\rm coh}$
CoMnVAl	Theory, this work	5.75	216.73	4.86	19.53
	Other theory	5.74[a]			
	Expt.	5.81[b], 5.80[c], 5.78[d], 5.75[e]			
CoMnVGa	Theory, this work	5.77	221.24	3.39	18.20
	Other theory	5.75[f]			
	Expt.	5.82[g]			

Comparison with previous calculation and experiments: [a]Refs. [32,33], [b]Ref. [29], [c]Ref. [39], [d]Ref. [40], [e]Ref. [41], [f]Ref. [36], [g]Ref. [42].

Compared with CoMnVAl, CoMnVGa possesses more electrons. These extra electrons from Ga atoms will fill the p-d hybridized bonding states of the 1nn Co–Ga and Mn–Ga atom pairs. This band filling will result in the increase of bulk modulus from 216.73 GPa in CoMnVAl to 221.24 GPa in CoMnVGa.[37] The pressure derivatives of bulk moduli are also listed in Table 1.

In solid, cohesive energy describes not only the strength of chemical bonding among the constituent atoms, but also the thermodynamic stability of compounds. Theoretically, the cohesive energy of CoMnVAl and CoMnVGa compounds in this work can be obtained by $E_{\rm coh} = (E^{\rm iso}_{\rm Co} + E^{\rm iso}_{\rm Mn} + E^{\rm iso}_{\rm V} + E^{\rm iso}_{\rm Al/Ga}) - E^{\rm Total}_{\rm CoMnVAl/Ga}$. Here, $E^{\rm iso}_{\rm Co}$, $E^{\rm iso}_{\rm Mn}$, $E^{\rm iso}_{\rm V}$ and $E^{\rm iso}_{\rm Al/Ga}$ are isolated atomic energy of Co, Mn, V, and Al/Ga atoms, respectively. $E^{\rm Total}_{\rm CoMnVAl/Ga}$ is the total energy of CoMnVAl/Ga compounds. The cohesive energy of CoMnVAl and CoMnVGa compounds are calculated to be 19.53 eV and 18.20 eV, respectively. Their positive cohesive energy values suggest that these two compounds are both thermodynamically stable. In contrast with CoMnVGa, CoMnVAl is more stable owing to higher cohesive energy. On the other hand, these high values mean strong chemical bonding in CoMnVAl and CoMnVGa compounds, which can be compared with other Fe-based Heusler compounds.[38] The slight difference of cohesive energy values between CoMnVAl and CoMnVGa suggests the distinct electrons distribution character and consequently the filling style of band structure, which affects their electronic structures and mechanical properties.

3.2. Electronic structure

In order to investigate the magnetism of CoMnVAl and CoMnVGa compounds at ground states, the total energy under non-magnetic, antiferromagnetic and ferromagnetic configuration was calculated at different unit cell volumes around the equilibrium unit cell volume. Then the calculated total energy was fitted with the Murnaghan's equation of state.[43,44] The obtained results for CoMnVGa and CoMnVAl are shown in Figs. 2(a) and 2(b), respectively. One can find that both CoMnVGa and CoMnVAl compounds show the lowest local minimum under non-magnetic configuration, which suggests that the most stable states of CoMnVGa and CoMnVAl compounds are non-magnetic. The fitted bulk modulus B and the equilibrium volume V are well consistent with the above calculations.

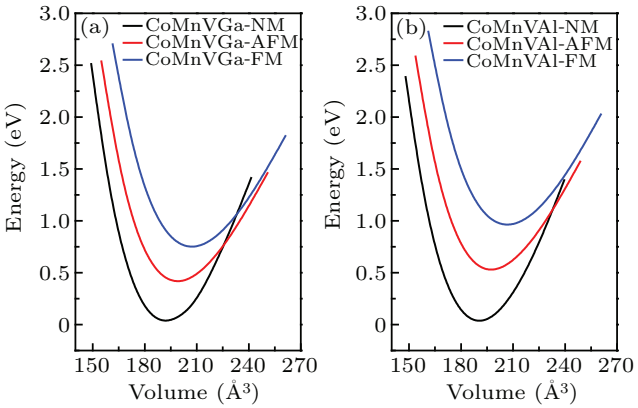


Fig. 2. Calculated total energy under non-magnetic, antiferromagnetic and ferromagnetic configuration for (a) CoMnVGa and (b) CoMnVAl as a function of unit cell volume. The minima of total energy at non-magnetic states were set to zero.

To confirm the non-magnetism in CoMnVAl and CoMnVGa compounds, electronic density of states (DOS) were calculated and shown in Fig. 3. In both CoMnVAl and CoMnVGa, electrons equally fill the 12 bands in spin up and spin down directions. This special band filling style leads to non-spin polarized states. Thus, at ground states, there are no finite magnetic moments for Co, Mn, V and Al\Ga atoms. This correspondingly results in zero total magnetic moments, agreeing with published observations.[13,29,31] To conclude, both CoMnVAl and CoMnVGa compounds are non-magnetic due to symmetric band filling of electrons.

Experimentally, the resistivity of CoMnVAl[39] and CoMnVGa[42] dependent temperature curves show kink behaviors around 200 K and 235 K, respectively. Above these kink points, small negative temperature coefficients of resistance were observed. It could be considered as the semiconductor-like behavior. From the calculated density of states in Fig. 3, one also finds that CoMnVAl and CoMnVGa compounds show semiconductor-like behavior. This character is similar to other equiatomic quaternary Heusler compounds with 24 valence electrons, i.e., CoFeTiAl, FeMnCrAl, FeMnVSi, FeCrVAs, FeMnTiAs, CoCrVSi, CoMnTiSi, and CoCrTiAs.[32] Although there are finite DOS near the Fermi level which depends on the employed computational approach,[16] the experimentally observed anomalous negative temperature dependent coefficients of electrical resistivity support semiconductor-like behaviors in CoMnVAl and CoMnVGa, similar as the well-known Fe_2VAl.[45] Thus, here we prefer to consider CoMnVAl and CoMnVGa as the non-magnetic semiconductor. Further experiments, such as high-resolution photoemission measurements, are necessary to confirm the semiconductor character of CoMnVAl and CoMnVGa compounds.

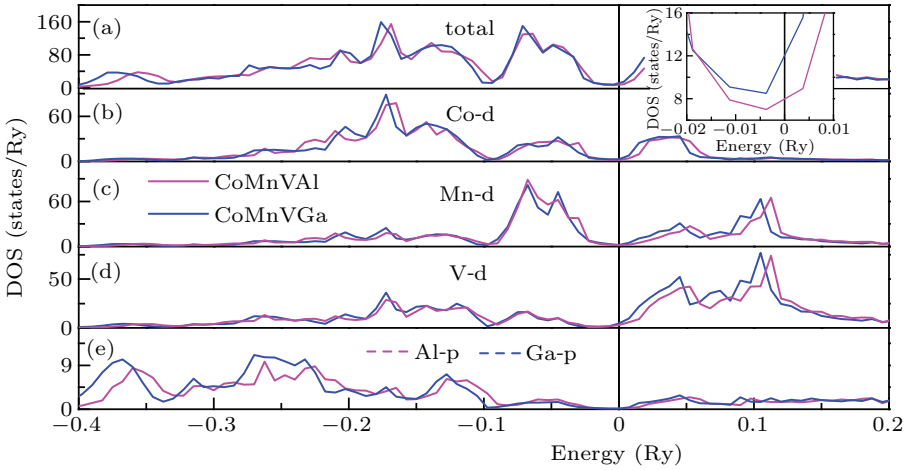


Fig. 3. Calculated electronic DOS plots for CoMnVAl and CoMnVGa compounds. The total DOS, the partial DOS for d-electrons of Co, Mn, V atoms and for p-electrons of Al\Ga atoms are illustrated in (a)–(e), respectively. The inset shows the enlarged total DOS distribution near the Fermi level. The pseudo-gap at Fermi level indicates the semiconductor-like behavior in CoMnVAl and CoMnVGa compounds.

3.3. Lattice dynamics properties

For CoMnVAl and CoMnVGa compounds, the calculated phonon dispersions along the selected high symmetry lines, together with corresponding total and partial density of phonon states (DOPS) at ground state are shown in Figs. 4(a)–4(d). For Heusler compounds, there are four atoms in primitive cell. Theoretically, in phonon dispersions, the optical and acoustic modes are totally twelve for any wave vector. Nine branches are optical modes, which distribute at high frequency zone. The left three phonon modes at low frequency are acoustic. Actually, as shown in Figs. 4(a) and 4(c), the number of branches may be reduced owing to the degeneracy along certain high symmetry directions. For CoMnVGa, its optical phonons mainly distribute from 5.26 THz to 9.40 THz in dispersion. And the optical phonon modes are coupled together. For CoMnVAl, it is another case. Its three optical phonon modes at high frequency range separate from other optical branches at lower frequency part, resulting in the obvious gap. From the DOPS, one can find the calculated gap is about 1.45 THz. By analyzing the partial DOPS of CoMnVAl, we find the separated three optical phonon modes at higher frequency range are mainly from Al atom with lower mass.

For CoMnVGa, due to the similar atomic masses of Co, Mn, V and Ga, its phonon branches are coupled together and thus no gap was observed.

In phonon dispersions of these two compounds, no any imaginary phonon frequencies were observed throughout the whole Brillouin zone. This means that CoMnVAl and CoMnVGa compounds are both dynamically stable in LiMgPdSn-type structure. However, in Figs. 4(a) and 4(c), their two transverse acoustic phonon modes (TA_1 and TA_2) at low frequency part show different change behaviors. For CoMnVGa, its TA modes become softing along *G*–*X*–*W* and *L*–*W* directions while in CoMnVAl these modes are hard without softing tendency. Thus, CoMnVAl is more stable at ground state without any possibility of dynamical instability.

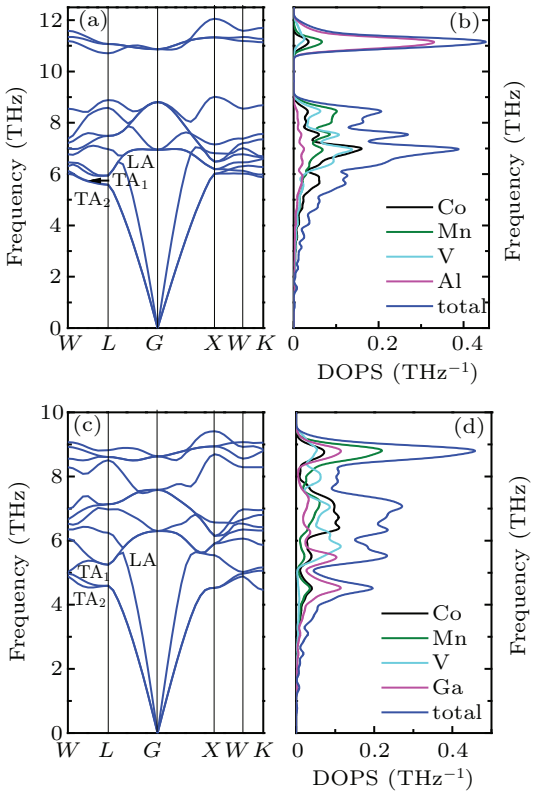


Fig. 4. The calculated phonon dispersions of (a) CoMnVAl and (c) CoMnVGa in LiMgPdSn-type structure. The high symmetry *q* point path in the Brillouin zone is chosen as $W(0.5, 0.25, 0.75) \rightarrow L(0.5, 0.5, 0.5) \rightarrow G(0, 0, 0) \rightarrow X(0.5, 0, 0.5) \rightarrow W(0.5, 0.25, 0.75) \rightarrow K(0.375, 0.375, 0.375)$. LA and TA represent longitudinal and transverse acoustic phonon modes. No imaginary frequencies of unstable modes are observed in the whole frequency space. Site projected density of phonon states of CoMnVAl and CoMnVGa are shown in (b) and (d), respectively, showing the distribution of amplitudes of atoms in the frequency range.

In phonon dispersions, two transverse acoustic phonon modes and one longitudinal acoustic phonon mode (LA) closely relate to the elastic parameters C_{44}, $C' = \frac{C_{11}-C_{12}}{2}$ and $C_{\mathrm{L}} = \frac{C_{11}+C_{12}+2C_{44}}{2}$.[46] Thus, the stability of CoMnVAl and CoMnVGa compounds can also be defined from elastic parameters. Moreover, the mechanical properties can be directly derived from elastic parameters, helping identify the possibility of future technical application. They will be discussed detailedly in the following section.

3.4. Elastic properties

In solids, the mechanical behaviors are defined by constitutive relations between stress σ_{ij} and strain ε_{kl}. Within Hook's law, their relations can be expressed as $\sigma_{ij} = \sum_{k,l=1}^{3} C_{ijkl}\varepsilon_{kl}$, where i, j, k and l are indices running from 1 to 3. Here C_{ijkl} is defined as elastic constant and used to describe the elastic properties of materials. It is a fourth-rank elasticity tensor with $3^4 = 81$ components. Due to the symmetry $C_{ijkl} = C_{klij} = C_{jikl} = C_{ijlk}$, the number of independent elastic constant reduced to at most 21.[26,47] They can be arranged in a symmetric 6×6 matrix. Adopting Voigt notation, the above $\sigma_{ij} = \sum_{k,l=1}^{3} C_{ijkl}\varepsilon_{kl}$ can be rewritten as $\sigma_{\alpha} = \sum_{\beta=1}^{6} C_{\alpha\beta}\varepsilon_{\beta}$, where the new α and β indices run from 1 to 6. In the Voigt notation the pair of indices ij are replaced by index α according to: $\alpha = 1, 2, 3$ for $ij = 11, 22, 33$; $\alpha = 4$ for $ij = 23$ or 32; $\alpha = 5$ for $ij = 13$ or 31 and $\alpha = 6$ for $ij = 12$ or 21, respectively. The number of elastic constants can be further reduced due to crystal symmetry. In cubic crystals, under symmetry there are the following conditions: $C_{11} = C_{22} = C_{33}$, $C_{12} = C_{23} = C_{13}$ and $C_{44} = C_{55} = C_{66}$. Thus, there are only three independent elastic constants C_{11}, C_{12} and C_{44}. In this work, the method in Ref. [26] was adopted to calculate C_{11}, C_{12} and C_{44}, whose effectiveness in metals and alloys has been proved by numerous works.[22–25]

To further investigate the phase stability and mechanical properties, the elastic constants of CoMnVAl and CoMnVGa compounds were calculated and shown in Table 2. As mentioned in previous part, CoMnVAl and CoMnVGa compounds both state in LiMgPdSn-type cubic structures. Thus, they have three independent elastic constants, which are C_{11}, C_{12} and C_{44}, respectively. For cubic compounds, the necessary and sufficient mechanical stability conditions are given by (1) $C_{11} > 0$; (2) $C_{44} > 0$; (3) $C_{11} + 2C_{12} > 0$; (4) $C' = \frac{C_{11}-C_{12}}{2} > 0$.[48] From the summarized elastic constants in Table 2, one can firstly find that CoMnVAl and CoMnVGa compounds both satisfy the necessary and sufficient criteria of mechanical stability very well. Comparatively, CoMnVAl compound possesses higher values of four mechanical stability conditions, indicating a more stable state. Secondly, based on elastic constants in Table 2, elastic constants C_{L} of CoMnVAl and CoMnVGa compounds are calculated to be 385.6 GPa and 376.4 GPa, respectively. Thus, combining their positive C_{44} and C', one can conclude that their three branches of acoustic modes, i.e., TA_1, TA_2 and LA, should lie in positive frequency range. Consistent with the phonon dispersions of both CoMnVAl and CoMnVGa compounds (see Fig. 4), no imaginary phonon frequencies were observed in the whole Brillouin zone at ground states. Comparatively, higher values of C_{44} and C' in CoMnVAl means harder TA modes in phonon dispersions than those of CoMnVGa. Thus, no tendency of modes softing occurs in phonon dispersions of CoMnVAl as shown in

Fig. 4(a). Additionally, for cubic Heusler compounds, when the Zener anisotropy factor A ($A = \frac{2C_{44}}{C_{11}-C_{12}} = \frac{C_{44}}{C'}$)[49] deviates from 1, the cubic compounds can also become unstable. Especially when the value of A is above 2, the cubic compounds will transform to low symmetric martensitic phases.[50] In this work, the Zener anisotropy factor A of the CoMnVAl and CoMnVGa compounds are 1.00 and 1.27, respectively. CoMnVAl is perfectly isotropic, ensuring a more stable cubic structure. Thus, from the point view of phonon dispersion, cohesive energy, elastic constants and Zener anisotropy factor, more stable phase stability was verified in CoMnVAl compound.

Table 2. Calculated elastic constants C_{ij}, C',Cauchy's pressure C_p, Zener anisotropy factor (A), shear modulus (G), $(B/G)(G/B)$ ratio, young's modulus (E), Poisson's ratio (υ), Vickers hardness (H_V) and melting temperature (T_m) of CoMnVAl and CoMnVGa compounds.

	Unit	CoMnVAl	CoMnVGa
C_{11}	GPa	386.12	350.56
C_{12}	GPa	132.04	156.58
C_{44}	GPa	126.51	122.82
C'	GPa	127.04	96.99
C_p	GPa	5.53	33.76
A		1.00	1.27
G_V	GPa	126.72	112.49
G_R	GPa	126.72	111.00
G_H	GPa	126.72	111.74
$(B/G_H)(G_H/B)$		1.71 (0.58)	1.97 (0.51)
E	GPa	318.16	286.92
υ		0.255	0.28
H_V	GPa	15.40	11.93
T_m	K	2835.34 ± 300	2625.16 ± 300

Based on the elastic constants of single crystal, elastic moduli can be derived by widely used Voigt–Reuss–Hill (VRH) approximation.[51–53] According to Voigt and Reuss approximations, the bulk and shear moduli of cubic Heusler compounds are deduced by the equations

$$B_V = B_R = \frac{C_{11}+2C_{12}}{3}, \quad G_V = \frac{(C_{11}-C_{12}+3C_{44})}{5},$$
$$G_R = \frac{5(C_{11}-C_{12})C_{44}}{4C_{44}+3(C_{11}-C_{12})}.$$

By VRH approximation, i.e., $B_H = \frac{B_V+B_R}{2}$ and $G_H = \frac{G_V+G_R}{2}$, average bulk and shear moduli can be obtained. Furthermore, the Young's modulus E and Poisson's ratio υ can be derived from $E = \frac{9BG}{3B+G}$ and $\nu = \frac{3B-2G}{2(3B+G)}$, respectively.[26] The calculated values are listed in Table 2.

The claim about mechanical properties of CoMnVAl and CoMnVGa compounds can be directly supported by obtained elastic moduli and their derivative parameters. Based on Pugh's work,[54] a simple relation between ductile properties and elastic moduli was proposed. It is well known that the bulk modulus B describes the resistant to fracture while the shear modulus G represents the resistance to plastic deformation. A B/G ratio can be used to distinguish the ductile and brittle materials. Generally, the higher B/G ratio means more ductile nature, whereas the lower ratio corresponds to brittle character. A critical ratio to separate ductile and brittle material is about 1.75. For CoMnVGa compound, its B/G ratio is 1.98, which is higher than the critical value. Thus, CoMnVGa can be defined as the ductile material. While for CoMnVAl, the B/G ratio (= 1.71) is closed to the critical value, which indicates a boundary state combining almost equal contribution from ductility and brittleness. Similarly, Poisson ratio, υ, has been considered as another criterion to classify the ductile and brittle materials. Generally, materials with υ above $1/3$ are ductile, whereas those with υ below $1/3$ are brittle.[55] For CoMnVAl and CoMnVGa in this work, their Poisson ratios are both below $1/3$. Thus, they are both brittle materials from the point of Poisson ratio. The different classifications based on these two criteria just reveal their imprecision in nature sometime. It should be noted that the critical value of Poisson ratio is not given explicitly in Ref. [55]. It is just based on properties reported for the materials tabulated by Frantsevich and was later accepted as empirical rule.[56] Although the classification is imprecise, both B/G ratio and Poisson ratio of CoMnVGa are higher than those of CoMnVAl, meaning better ductile properties in CoMnVGa.

Vickers hardness is another parameter indexing mechanical performance, which is defined as the resistance of materials to indentation. It can be directly derived from elastic moduli in Table 2. Here the method developed by Tian et al., $H_V = 0.92k^{1.137}G^{0.708}$ [57] was adopted, in which k is G/B ratio. The calculated Vickers hardness for CoMnVAl and CoMnVGa are 15.4 GPa and 11.93 GPa, respectively. Generally, the boundary hardness to separate hard and soft materials is 10 GPa. Materials with hardness below 10 GPa are soft. Otherwise, they are hard.[58] Thus, both CoMnVGa and CoMnVAl are hard compounds. In this work, higher G/B ratio and G in CoMnVAl promote its Vickers hardness up to 15.4 GPa, indicating harder nature than CoMnVGa.

Thermodynamically, the strength of covalent bonding in solids can be quantificationally characterized by Debye temperature (Θ_D). Generally, the higher Θ_D corresponds to the stronger strength of covalent bonding.[59] For the current CoMnVAl and CoMnVGa compounds, their Θ_D were evaluated from the average sound velocity, which is based on the mechanical moduli and shown as below. The equation is given as $\Theta_D = \frac{h}{k_B}\left[\frac{3q}{4\pi}\frac{N_A\rho}{M}\right]^{1/3} \nu_m$,[60] where h and K_B are the Planch and Boltzmann constants, q is the number of atoms in each formula unit (Here $q = 4$ for CoMnVAl/Ga Heusler compounds), N_A is the Avogadro number, M is the molecular weight per formula unit, and ρ is the density. ν_m is the average velocity, expressed as $\frac{3}{\nu_m^3} = \frac{1}{\nu_L^3} + \frac{2}{\nu_T^3}$. Among it, ν_L ($\rho\nu_L^2 = B + \frac{4}{3}G$) and ν_T ($\rho\nu_T^2 = G$) are longitudinal and transverse sound velocities, respectively.[26] The obtained ν_L, ν_T, ν_m and Θ_D are

listed in Table 3. For CoMnVAl and CoMnVGa, their Θ_D are 630.32 K and 537.77 K, respectively. Stronger covalent bonding in CoMnVAl was confirmed by higher Θ_D. This relatively stronger covalent bonding in CoMnVAl will promote chemical bonds more directional and make it much brittler and harder. This claim agrees with the observed lower B/G ratio, Poisson's ratio, Cauchy pressure and higher Vickers hardness.

Table 3. Calculated longitudinal velocity, transverse velocity and average velocity (short as v_L, v_T and v_m, respectively) in m/s, and the Debye temperature (Θ_D) in Kelvin for CoMnVAl and CoMnVGa compounds at theoretical equilibrium states.

Compounds	v_L	v_T	v_m	Θ_D
CoMnVAl	7596.95	4348.80	4831.03	630.32
CoMnVGa	6747.13	3706.74	4131.98	537.77

The thermodynamic properties of CoMnVAl and CoMnVGa compounds can be also discussed from the melting point. Empirically, the melting points are evaluated by the equation of $T_m = (553 + 5.911C_{11}) \pm 300$[61] and listed in Table 2. The higher melting point values mean these two compounds are stronger against temperature. And it implies that properties at ground state mentioned above will be able to be preserved as much as possible at high temperature. The stable functional parameters are important for the future technical applications.

4. Discussion

4.1. Mechanical properties

To help understand different mechanical properties of CoMnVAl and CoMnVGa compounds, further investigations have been carried out to reveal their physical origin in the following part. Firstly, the Cauchy pressure ($C_p = C_{12} - C_{44}$) in solid reflects the bonding character. According to Pettifor,[62] negative C_p means more covalent character of bonding while the larger positive one shows more metal bonding character. In this work, C_p values of CoMnVGa and CoMnVAl are 33.76 GPa and 5.53 GPa, respectively. Obviously, CoMnVGa shows stronger metal bonding nature. The near zero C_p value in CoMnVAl reflects weaker metallic bonding compensated by the increase of covalent bonding component. Secondly and qualitatively, the atomic bonding character can be directly revealed from the contour plot of electron localization function in (110) plane, as shown in Fig. 5(a) for CoMnVAl and (b) for CoMnVGa. One can find that charge accumulation level between 1nn Co–Al and Mn–Al atom pairs in CoMnVAl is higher than 1nn Co–Ga and Mn–Ga atom pairs in CoMnVGa. This results in higher covalent bonding contribution and consequently weaker metallic properties in CoMnVAl. Thirdly and quantitatively, based on the quantum theory of atoms in molecules suggested by Bader and Henkelman,[63,64] the charge-density topology can help quantify the charge distribution in both CoMnVAl and CoMnVGa compounds. Based on this method, the covalent character of CoMnVAl and CoMnVGa can be also identified by calculating the charge on specific atoms in their sub-lattice. They are $Co^{-0.34}(Mn^{0.06}V^{0.24}Al^{0.04})$ and $(Co^{-0.40}Mn^{-0.12}V^{-0.18})Ga^{0.70}$, respectively. Comparatively, CoMnVAl shows relatively higher covalent character than CoMnVGa. Moreover and physically, the calculated density of states can further help understand the origin of different mechanical properties. Generally, the higher density of states values near the Fermi level, the stronger metallic bonding. Here CoMnVGa possesses higher DOS values near the Fermi level, consistent with better metallic properties. For CoMnVAl, its DOS values near the Fermi level is slightly decreased when compared with CoMnVGa, which means the level of covalent component is enhanced. Thus, CoMnVAl shifts to a coexistence state of covalent character and metallicity, which agrees with the near zero Cauchy pressure. The increased covalent component in CoMnVAl is responsible for its relatively weak ductility. Due to the intrinsic character of covalent bonding, the enhanced covalent hybridization level in CoMnVAl also leads to higher mechanical strength and Vickers hardness.

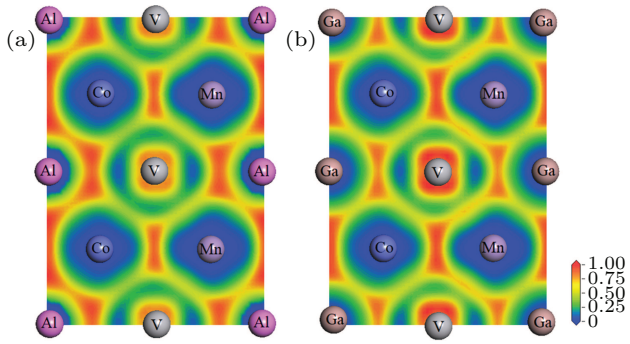


Fig. 5. The electron localization function distribution on (110) plane of (a) CoMnVAl and (b) CoMnVGa in LiMgPdSn-type structure. The reference bar for electron localization function value is provided on the bottom right corner. The higher electron accumulation level between 1nn Al and Co/Mn atom pairs in CoMnVAl illustrates stronger covalent bonding hybridization.

Here detailed analysis has proved stronger covalent bonding in CoMnVAl, dominated by p–d hybridization between main group element Al and transition metal atoms Co/Mn. In Heusler compounds, this kind of covalent hybridization not only contributes to functional properties, but also determines lattice size. Experimental observations in cubic Co–Mn–Ga and tetragonal Ni–Mn–Ga–Cu Heusler compounds suggest that stronger covalent hybridization can shrink lattice constants.[34,35] Thus, except the intrinsic smaller atomic radius of Al, the contracted lattice size of CoMnVAl in this work should also owe to stronger covalent hybridization. Furthermore, stronger interatomic covalent hybridization gives rise of higher cohesive energy (see Table 1) and results in more thermodynamically stable structure. To summarize, the chemical bonding between constituent 1nn atom pairs, especially which

between transition metals and p-block elements, determines the mechanical properties of Heusler compounds. Thus, tuning the chemical bonding between 1nn atom pairs by doping elements will be an effective way to improve their mechanical properties for technical application. Thus, in Heusler compounds, the rich source of constituent elements in period table provides a wide platform to tune their mechanical properties.

Si and Ge are widely used semiconductors as substrate to grow multiple functional film samples. Due to the nature of covalent bonding, their negative Cauchy pressure and lower B/G values make them brittle.[65,66] Here in non-magnetic semiconductor CoMnVAl, the increased metal bonding component raises the Cauchy pressure slightly above zero (5.53 GPa) and results in relatively higher B/G value than those of Si and Ge. This enhances the ductility of CoMnVAl and keeps its strength at relatively high level. Consequently, the balance of covalent bonding and metal bonding in CoMnVAl could lead to better comprehensive mechanical properties. Additionally, based on the available data,[65] we find the Vickers hardness of CoMnVAl can be compared with Si and Ge. Importantly, the isotropic structure in CoMnVAl, i.e., $A = 1$, will benefit to grow high quality samples avoiding defects or micro-cracks.[67] Thus, non-magnetic semiconductor CoMnVAl would be more attractive working as substrate material servicing in spintronic device.

Currently, isostructural Heusler compounds Co_2MnGa and Co_2MnAl are predicted as magnetic Weyl semimetals.[7–10] To grow high quality film samples, the suitable non-magnetic semiconductor materials as substrate are necessary. Here, CoMnVAl is considered as non-magnetic semiconductor. Its lattice parameters (5.75 Å) is close to those of Co_2MnAl (5.75 Å)[9] and Co_2MnGa (5.77 Å).[10] Combining the stable crystal structure and better comprehensive mechanical properties mentioned in previous part, the less lattice mismatch and similar structure can fulfill the criteria of structural and chemical compatibilities at the interface, which makes CoMnVAl and magnetic topological semimetal Co_2MnAl/Ga suitable to be grown in heterostructures for spintronic application. As discussed in the following section, atomic disordered occupation can activate magnetism and destroy the semiconductor property. Thus, to realize the semiconductor character of CoMnVAl practically, effective experiments are necessary to ensure the forming of high ordered Heusler structure. We hope that current work can provide more options or suggestions for future spintronic applications.

4.2. Origin of vanished magnetism in CoMnVAl/Ga

For CoMnVAl and CoMnVGa compounds, there are 24 valence electrons in each primitive cell. Based on the well-known Slater–Pauling rule,[14,15] their total magnetic moments should reach zero. Considering the constituent atoms of Co, Mn and V, these two compounds can be antiferromagnetic, fully compensated ferrimagnetic or non-magnetic. Due to the fact that Heusler alloys cannot fulfill the symmetry requirements needed to be antiferromagnets, these two compounds are not normal antiferromagnets even though showing vanished moments.[18] Thus, CoMnVAl and CoMnVGa can be non-magnetic or fully compensated ferrimagnetic. Sometimes one may be confused on the fully compensated ferrimagnetic and antiferromagnetic compounds. In common, they both not only possess finite atomic magnetic moments, but also their atomic magnetic moments are antiparallelly aligned and compensate each other, resulting in zero total moment. However, magnetic exchange interactions are strong in compensated ferrimagnetic compounds, which has been confirmed by experimentally observed high Curie temperature.[17]

Comparing with the compensated ferrimagnetic compounds, the vanished atomic magnetic moments in non-magnetic Heusler compounds were considered to stem from the atomic preferential occupation. According to valence electron rule to predict the half-metallic compensated ferrimagnetic behavior in Heusler compounds, even partial occupation of B sites by localized atoms such as Mn is sufficient to enforce the formation of localized magnetic moments on these sites.[18] Otherwise, it will be non-magnetic. The validity of this rule has been verified from both theory and experiment in the case of $CoMn_{1+x}V_{1-x}Al$[31] and $Mn_{2-x}Fe_xMn_{1-x}V_xAl$[17] systems. In Heusler family, Co_2VAl/Ga and Mn_2VAl/Ga are isostructural compounds with $L2_1$ structure. In primitive cell, two Mn or Co atoms occupy A/C sites while V and Al/Ga atoms state in B and D sites, respectively. By alloying equal proportion of Co_2VAl/Ga and Mn_2VAl/Ga, LiMgPdSn-type equiatomic quaternary CoMnVAl/Ga can be obtained. Naturally, in CoMnVAl and CoMnVGa compounds, localized Mn atoms lie in A or C site, not the B/D sites. Under this atomic configuration, the magnetic exchange coupling can not be established and thus induce the formation of magnetic moments. Thus, CoMnVAl and CoMnVGa compounds are both non-magnetic theoretically.

Experimentally, the observed magnetic moments of CoMnVAl and CoMnVGa compounds are 1.09 μ_B/f.u.[41] and 0.37 μ_B/f.u.,[42] respectively. They both deviate from the zero value. These differences between experiment and theory were considered to be from atomic disorder occupation or certain variation of composition. For CoMnVAl film,[41] experiments reveal that its constituent atoms become disordered due to the insufficient heat treatment temperature because of Mn sublimation. Except the original A/C sites, localized Mn atoms can also occupy B and/or D sites and were supposed to become preferentially the nearest neighbors of Co atoms. This atomic configuration gives rise of non-zero magnetic moment. For nominal CoMnVGa bulk sample,[42] the experimental atomic

percent of Co, Mn, V and Ga are 25.7%, 26.5%, 20.7% and 26.9%, respectively. The atomic ratio is not equal any more. Extra Co and localized Mn atoms have to occupy the vacancy on B sites. Under this atomic configuration, the magnetic exchange interactions were established and induced the formation of magnetic moments. Here experimental observations reveal that magnetism can be activated when localized Mn atoms occupy B and/or D sites.

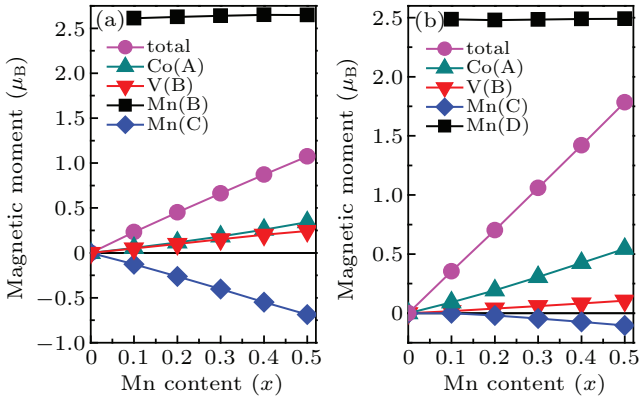


Fig. 6. The calculated total magnetic moments and atomic magnetic moments of (a) $CoMn_{1+x}V_{1-x}Ga$ and (b) $CoMn_{1+x}VGa_{1-x}$ ($x = 0$–0.5).

Inspired by the above experimental observations in the CoMnVGa and CoMnVAl compounds, two series compound of $CoMn_{1+x}V_{1-x}Ga$ and $CoMn_{1+x}VGa_{1-x}$ were considered here to further investigate the origin of vanished magnetism in CoMnVAl/Ga compounds. Based on atomic preferential occupation rule in Heusler compounds, the newly introduced Mn atoms will occupy the vacancy on B sites in $CoMn_{1+x}V_{1-x}Ga$ and D sites in $CoMn_{1+x}VGa_{1-x}$, respectively. Their total and atomic magnetic moments were calculated and the results are shown in Figs. 6(a) and 6(b). With increasing the content of Mn in these two series compound, the absolute values of the moments of Co(A), V(B) and Mn(C) all increase. The strongest and stable atomic magnetic moments were observed in Mn atoms on B and D sites. Although the magnetic moments of Mn on C site are antiparallel to other atomic moments, its negative contribution to total moment was compensated by moments on A, B and/or D sites, especially Mn atoms on B and D sites. Consequently, the total moments in both series gradually increase when Mn atoms were introduced to B and D sites. Similar results were obtained in $CoMn_{1+x}V_{1-x}Al$ and $CoMn_{1+x}VAl_{1-x}$ compounds. Thus, current calculations confirm that the magnetic exchange coupling can be activated when Mn atoms occupy B and D sites and magnetism will be induced. By analyzing the above theoretical findings and the available experiments, one can conclude that the formation of non-zero magnetic moments in CoMnVAl/Ga compounds result from the localized Mn atoms occupying B or D sites.

Since A and C sites are equivalent in $L2_1$-type Heusler structure, the magnetic moments of Mn on A sites will be parallel to those on C sites. Thus, if further introducing Mn on A sites in $CoMn_{1+x}V_{1-x}Ga$ and $CoMn_{1+x}VGa_{1-x}$ compounds, the increased negative contribution from magnetic moments of Mn on A and C sites will decrease the total moments. Based on Slater–Pauling rule, the total moment of $Co_{1-y}Mn_{1+x+y}V_{1-x}Ga$ and $Co_{1-y}Mn_{1+x+y}VGa_{1-x}$ compounds can reach zero if keeping the total valence electrons of 24 in each primitive cell. Following the above calculation and analysis, novel fully compensated ferrimagnetic compounds $Co_{1-x}Mn_{1+2x}V_{1-x}Ga$ and $Co_{1-2x}Mn_{1+3x}VGa_{1-x}$ were theoretically designed. The calculated total magnetic moments and atomic magnetic moment of $Co_{1-x}Mn_{1+2x}V_{1-x}Ga$ and $Co_{1-2x}Mn_{1+3x}VGa_{1-x}$ were shown in Fig. 7. As expected, atomic moments of Mn on A and C sites are both negative and antiparallel to atomic moments of Co(A), V(B) and Mn(B). Due to the competition and compensation between antiparallel aligned atomic moments, the near zero total moments were realized in both series compound as expected. Thus, the fully compensated ferrimagnetic compounds are theoretically achieved in off-stoichiometric $Co_{1-x}Mn_{1+2x}V_{1-x}Ga$ and $Co_{1-2x}Mn_{1+3x}VGa_{1-x}$ system. Our recent experimental work finds that these compounds also possess high Curie temperature and spin polarization, which makes them attracting candidate materials for spintronic applications.[68] Similar results were obtained in off-stoichiometric Co–Mn–V–Al compounds. Our work suggests that the occupation of localized Mn atoms on B and/or D site can activate magnetism and help achieve the fully compensated ferrimagnetic compounds in Heusler compounds, which is not only restricted to B site as mentioned in valence electron rules.[18] Thus, our analysis enriches the original valence electron rules and provides more choices to search for fully compensated ferrimagnetic materials in Heusler family.

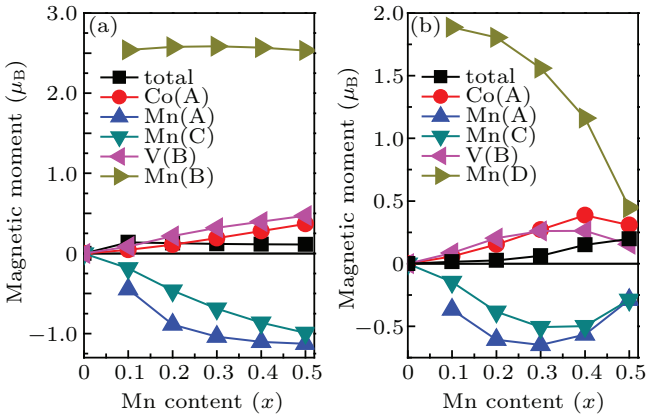


Fig. 7. The calculated total magnetic moments and atomic magnetic moments of $Co_{1-x}Mn_{1+2x}V_{1-x}Ga$ and $Co_{1-2x}Mn_{1+3x}VGa_{1-x}$ ($x = 0$–0.5) with total valence electrons of 24 in primitive cell.

5. Conclusion

In this work, crystal structure, electronic structure, lattice dynamics, elastic properties and the origin of vanished

magnetism in non-magnetic Heusler compounds CoMnVZ (Z = Al, Ga) were studied by the first principle calculations. Owing to the similar constituent atoms, both CoMnVAl and CoMnVGa compounds are stable in LiMgPdSn-type structure with similar lattice size, phonon dispersions and electronic structures. Due to the atomic preferential occupation, localized atoms Mn occupy C sites rather than B or D sites in both CoMnVAl and CoMnVGa, which leads to symmetric band filling style and consequently makes them the most stable in non-magnetic states. Furthermore, in CoMnVAl and CoMnVGa, the experimental negative temperature dependent coefficient of resistance and theoretical density of states character near Fermi level indicate their semiconductor character. However, by contrast, we find that CoMnVAl is more stable in LiMgPdSn-type structure than isostructural CoMnVGa. Qualitative and quantitative analysis reveals that the increased component of directional covalent bonding in CoMnVAl, especially those between 1nn transition metals and p-block elements, enhances its mechanical strength and Vickers hardness compensated with slightly decreased ductility, which consequently results in better comprehensive mechanical properties. Furthermore, practically and importantly, structural and chemical compatibilities at the interface make non-magnetic semiconductor CoMnVAl and magnetic topological semimetal Co_2MnAl/Ga suitable to be grown in heterostructures for spintronic application. Importantly, we find that tuning localized atoms Mn to occupy B or/and D sites can activate magnetic exchange interactions and induce the formation of magnetism. Immediately afterwards, based on Slater-Pauling rule, newly compensated ferrimagnetic compounds were achieved in off-stoichiometric Co–Mn–V–Al/Ga compounds with the total valence electrons of 24, which makes them more attractive for spintronic research. We hope that our work will provide more choices for spintronic application.

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