

# First-principles study of magnetic and spin-dependent transport properties of $\text{Mn}_2\text{VZ}$ ( $Z = \text{Al}, \text{Ga}$ ) with negative spin polarization using a disordered local moment approach at finite temperatures

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First-principles studies were performed on two Mn-based ferrimagnetic Heusler compounds with  $L2_1$  and  $B2$  structures, that is,  $\text{Mn}_2\text{VZ}$  ( $Z = \text{Al}$  or  $\text{Ga}$ ). The aim was to investigate their magnetic properties, electronic structures, and spin-resolved longitudinal conductivity at both zero and elevated temperatures. Density functional theory and functional integral theory were used. This approach incorporates transverse spin fluctuations through a disordered local moment method and the coherent potential approximation. In most cases, the calculated theoretical Curie temperatures were lower than the experimental values. Alloys with a  $B2$  structure exhibit higher Curie temperatures compared to compounds with an  $L2_1$  structure. Calculations of the temperature dependence of the density of states (DOS) indicate that the half-metallic electronic structure collapses due to self-energy effects that reflect spin disorder at nonzero temperature. In other words, transverse spin fluctuations are renormalized in the electronic structure. However, the spin-resolved longitudinal conductivities demonstrated an improved spin polarization, particularly for  $\text{Mn}_2\text{VGa}$  with an  $L2_1$  structure. The competition between the half-metal-to-normal-metal transitions, which are caused by a modification of the DOS, and scattering coming from spin-disorder explains this behavior of conductivities. Both of these effects are induced by transverse spin fluctuations. Additionally, the results show that the spin polarizations defined in terms of the conductivities and of the DOS do not follow the same trends at  $T > 0$ . Their temperature dependencies are not the same, particularly for the  $L2_1$  structures, although they are related to each other. Finally, the total energy landscape of the paramagnetic state was calculated using the fixed spin moment method to investigate the strength of the longitudinal spin fluctuations. These results suggest that the alloys may exhibit strong longitudinal spin fluctuations.

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## I. INTRODUCTION

Half-metallic Heusler alloys, such as  $\text{Co}_2$ -based Heusler alloys [1–3], are essential materials in the field of spintronics, owing to their unique electronic structure. In half metallic materials, one spin channel is metallic, whereas the other spin channel is semiconducting. Several Mn-based Heusler alloys exhibit interesting properties. For example,  $\text{Mn}_2\text{CoAl}$  [4] is a spin-gapless semiconductor and  $\text{Mn}_{1.5}\text{V}_{0.5}\text{FeAl}$  [5,6] and  $\text{Mn}_2\text{Co}_{0.5}\text{V}_{0.5}\text{Z}$  ( $Z = \text{Al}$  or  $\text{Ga}$ ) [6–8] are fully compensated ferrimagnets that exhibit no net magnetization while maintaining a half-metallic electronic structure.

Among the Mn-based Heusler compounds it is known that  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  are half-metallic ferrimagnets that exhibit a negative spin polarization. Their Fermi levels are located in the band gap formed in their majority spin channel [9–13]. This means that the sign of spin polarization at the Fermi level is opposite to the bulk magnetization. According to the electron-counting rule based on the localized part of the Slater-Pauling curves, the number of valence electrons ( $n_v$ ) in the primitive cell of  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  is 22. This property results in an expected total magnetic moment of  $2 \mu_B$  [12]. This value is lower than that of  $\text{Co}_2$ -based half-metallic ferromagnetic Heusler compounds, which have more valence electrons. Examples include  $\text{Co}_2\text{MnSi}$  and  $\text{Co}_2\text{FeSi}$  with  $n_v = 29$  and  $30$ , respectively.

Because of their low magnetic moments, stray fields are expected to be suppressed. Reducing the critical current should improve magnetization switching with spin-transfer torque and minimize electronic power consumption. Recently, current-perpendicular-to-plane giant magnetoresistance devices were fabricated using  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$ . These devices exhibit negative magnetoresistance of  $-4.4\%$  for

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Mn<sub>2</sub>VAI [14] and  $-1.8\%$  for Mn<sub>2</sub>VGa [15] at room temperature, respectively. A negative spin polarization was confirmed for these alloys.

From a spintronics perspective, a theoretical understanding of the temperature dependence of material-specific properties is highly desirable. These properties include spin polarization and the influence of chemical disorder on the electronic structure and related transport properties. This knowledge will provide a fundamental understanding of Heusler compounds and alloys, and assist in the fabrication of superior spintronics devices.

Theoretical knowledge based on density functional theory (DFT) is mostly limited to zero temperature. Therefore, attempts have been made to extend this method to include nonzero temperature ( $T > 0$ ) effects. To study the magnetic properties of Heusler alloys at  $T > 0$ , Heisenberg-type spin Hamiltonians or their extensions are typically used, as in Refs. [16,17].

Previous theoretical studies also attempted to estimate the Curie temperatures of Mn<sub>2</sub>VAI and Mn<sub>2</sub>VGa based on the above concept. For example, Chico *et al.* [18] calculated the Curie temperature of these alloys using an effective classical Heisenberg model combined with the Liechtenstein-Katsnelson-Antropov-Gubanov formula [19] and a Monte Carlo method. They calculated exchange interactions with several exchange correlation potentials. Their results for both local spin density approximation (LSDA) and generalized gradient approximation (GGA) also underestimated the Curie temperatures as compared with the experimental results. In the other study, Sasioglu *et al.* [20] have calculated the Curie temperature of Mn<sub>2</sub>VAI using an effective classical Heisenberg-type spin Hamiltonian. They extracted exchange interactions using the frozen magnon approach and estimated the Curie temperature with the mean-field approximation. Kübler [21] used a different approach for calculating the Curie temperature of Mn<sub>2</sub>VAI using a formula, which is similar to the random phase approximation in the Heisenberg model. In this case, the exchange interactions were also obtained from the spin-spiral excitation energies. However, results also underestimated the experimental values.

As several references have indicated,  $d$  electrons in metallic systems can exhibit both itinerant and localized properties [22,23]. For Mn<sub>2</sub>VAI, x-ray spectroscopy confirmed the itinerant nature of  $d$  electrons at the Mn atoms and the localized nature of those at the V atoms [24,25]. A similar situation is expected in the isostructural Mn<sub>2</sub>VGa. Therefore, a theoretical approach that incorporates both properties, not only localized picture but also itinerant picture, is necessary to describe metallic magnetic materials, including Mn-based Heusler alloys, at  $T > 0$ . Based on this concept, theoretical schemes for treating spin fluctuations in metallic magnets have been developed using the Hubbard model (see Refs. [26–31]). These schemes have been translated into first-principles methods (see Refs. [32–36]).

The aim of this study was to examine the temperature-dependent magnetic properties, electronic structures, and spin-resolved longitudinal conductivities of Mn<sub>2</sub>VAI and Mn<sub>2</sub>VGa based on spin fluctuation theory for metallic systems. The finite-temperature properties were calculated using functional integral theory based on DFT. This the-

ory expresses the transverse spin fluctuations at  $T > 0$  and incorporates the itinerant and localized properties of  $d$  electrons. The disordered local moment (DLM) method was employed to address transverse spin fluctuations at  $T > 0$  (see Refs. [32–36]). The spin-disordered electronic structure of the systems at  $T > 0$  was evaluated using coherent potential approximation (CPA). Using this scheme, we can calculate the temperature dependence of the electronic structure and transport properties from the electronic structures based on the DFT, and the linear response theory. Accordingly, the temperature dependence of the magnetization, DOS, and spin-resolved longitudinal conductivities of Mn<sub>2</sub>VAI and Mn<sub>2</sub>VGa was calculated for both the  $L2_1$  and  $B2$  structures.

Finally, the strength of the longitudinal spin fluctuations was explored by investigating the total energy landscape in paramagnetic states. The aim was to briefly study the properties of spin fluctuations in both alloys. The findings of this study clarify how transverse spin fluctuations alter the electronic structure and affect spin-resolved conductivity at  $T > 0$ .

## II. DETAILS OF THE CALCULATIONS

First-principles calculations based on DFT were used to investigate the temperature dependence of the magnetization, electronic structure, and transport properties. The electronic structure at zero temperature was calculated using a computer program based on the Korringa-Kohn-Rostoker (KKR) method combined with the atomic sphere approximation (ASA) [37,38]. The program also provides the tight-binding linear muffin-tin orbital (TB-LMTO) method. In this work, we used the TB-LMTO method in the program to calculate the electronic structure and potential parameters. After obtaining the TB-LMTO potential parameters for Mn<sub>2</sub>VAI and Mn<sub>2</sub>VGa, functional integral theory based on the TB-LMTO method was applied [39–43]. By using functional integral theory, the probability of spin transverse fluctuations  $\omega_i(T, \mathbf{e}_i)$  at temperature  $T$  and site  $i$  within the single-site approximation can be calculated in combination with the electronic structure obtained with DFT. This theory incorporates transverse spin fluctuations at  $T > 0$  by using a vector-type DLM picture. The evaluation of the functional integrals relies on the force theorem. Therefore, the longitudinal spin fluctuations were neglected. The details of formulation of the functional integral theory to obtain  $\omega_i(T, \mathbf{e}_i)$  based on the TB-LMTO and KKR methods are summarized in Refs. [44–50] and in [51,52], respectively.

In this study, the LSDA was employed with the von Barth-Hedin-Janak functional as the exchange-correlation potential. The lattice constants were set to 0.5875 nm and 0.5905 nm for Mn<sub>2</sub>VAI [10] and Mn<sub>2</sub>VGa [53], respectively. Spin-orbit coupling in the valence bands was neglected. The structure constants were expanded up to  $l = 2$ . The  $3d$  electrons of Ga were treated as core states.

TB-LMTO Hamiltonian matrix defined with the local spin frame  $H^{\text{TB-LMTO}}(\{\mathbf{e}\})$ , is given as follows:

$$H^{\text{TB-LMTO}}(\{\mathbf{e}\}) = C + \sqrt{\Delta} \tilde{S}(\{\mathbf{e}\})(1 - \gamma \tilde{S}(\{\mathbf{e}\}))^{-1} \sqrt{\Delta}. \quad (1)$$

$C$ ,  $\Delta$ , and  $\gamma$  are potential parameters of the LMTO method, which are diagonal matrices with respect to site, orbital, and

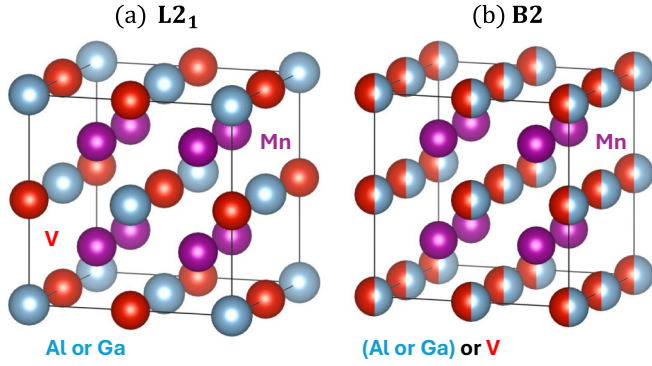


FIG. 1. Crystal structures of  $\text{Mn}_2\text{VAI}$  and  $\text{Mn}_2\text{VGa}$  with (a)  $L2_1$  and (b)  $B2$  structures.

spin indices. To express the transverse spin fluctuations, the bare structure constant matrix  $S$  is rotated by using the  $\text{SU}(2)$  rotation matrices  $U$ ,

$$\tilde{S}_{iL\sigma, jL'\sigma'}(\{\mathbf{e}\}) = \sum_{ss'} U_{\sigma s}(\mathbf{e}_i) S_{iLs, jL's'} U_{s'\sigma'}^\dagger(\mathbf{e}_j). \quad (2)$$

To obtain the TB-LMTO Hamiltonian defined with the global spin axis  $\tilde{H}^{\text{TB-LMTO}}$ , we also apply an  $\text{SU}(2)$  rotation matrix to the  $H^{\text{TB-LMTO}}$  as follows:

$$\tilde{H}_{iL\sigma, jL'\sigma'}^{\text{TB-LMTO}}(\{\mathbf{e}\}) = \sum_{ss'} U_{\sigma s}^\dagger(\mathbf{e}_i) H_{iLs, jL's'}^{\text{TB-LMTO}}(\mathbf{e}_i) U_{s'\sigma'}(\mathbf{e}_j). \quad (3)$$

Green's function including transverse spin fluctuations, defined with the global spin frame,  $G$  is given by

$$G(z, \{\mathbf{e}\}) = (z - \tilde{H}^{\text{TB-LMTO}}(\{\mathbf{e}\}))^{-1}. \quad (4)$$

More details to obtain Green's function including spin fluctuation and the way to treat the Green's function in the TB-LMTO method are found in Refs. [44–50]. Once we obtained the Green's function and  $\omega_i(T, \mathbf{e}_i)$ , the density of states (DOS)  $D(T, E)$  and the magnetization  $M_i(T)$  at site  $i$  and at  $T > 0$  are calculated by

$$D(T, E) = -\frac{1}{\pi} \text{Im} \left[ \text{Tr}_{iLs} \int d\mathbf{e}_i \omega_i(T, \mathbf{e}_i) G_{ii}(z^+, \mathbf{e}_i) \right], \quad (5)$$

$$M_i(T) = -\frac{1}{\pi} \text{Im} \left[ \text{Tr}_{iLs} \int_{-\infty}^{\infty} dE f(E, T, \mu) \times \int d\mathbf{e}_i \omega_i(T, \mathbf{e}_i) \sigma_z G_{ii}(z^+, \mathbf{e}_i) \right], \quad (6)$$

where  $G_{ii}$  is the on-site block of the Green's function  $G$  of the TB-LMTO method including transverse spin fluctuations.  $\sigma_z$  is the  $z$  component of the Pauli matrix.  $f$  is the Fermi-Dirac distribution. The trace is taken with respect to the space of the atom indices  $i$ , orbital indices  $L$ , and spin index  $s$ . The Kubo-Greenwood formula was used to calculate the spin-resolved conductivities:

$$\sigma_{\nu\nu}^s(T) = -\frac{\hbar}{4\pi V} \int_{-\infty}^{\infty} dE \left( -\frac{\partial f(E, T, \mu)}{\partial E} \right) \times \text{Tr}_{iL} \langle J_\nu (G(z^+, \{\mathbf{e}\}) - G(z^-, \{\mathbf{e}\})) J_\nu (G(z^+, \{\mathbf{e}\}) - G(z^-, \{\mathbf{e}\})) \rangle_{\omega_i(T, \mathbf{e}_i)}. \quad (7)$$

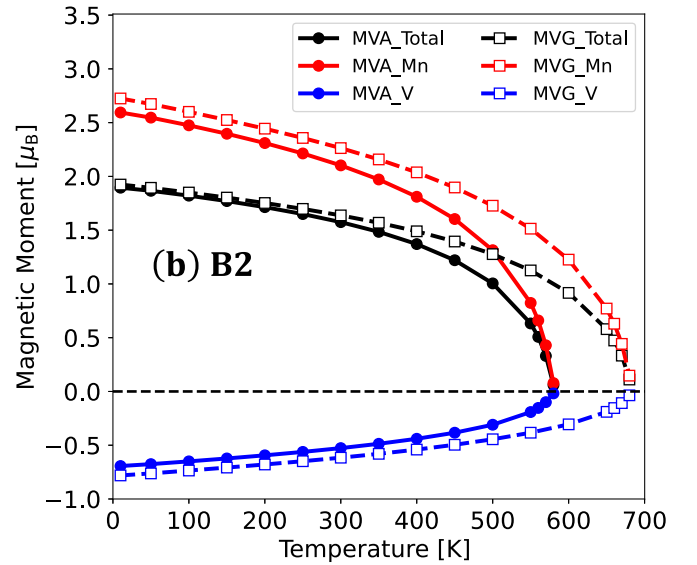
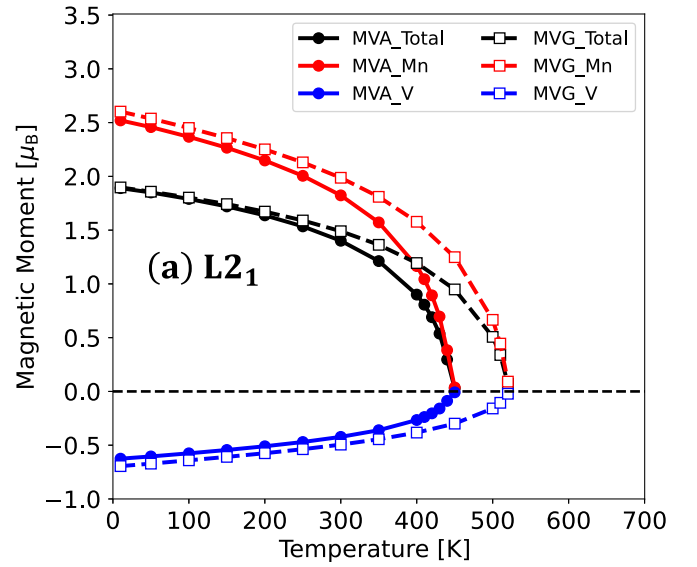


FIG. 2. Temperature dependence of the total magnetization and site-resolved magnetic moments of the Mn and V sites of  $\text{Mn}_2\text{VAI}$  (MVA, closed circles) and  $\text{Mn}_2\text{VGa}$  (MVG, open squares) with (a)  $L2_1$  and (b)  $B2$  structures. Black, red, and blue lines correspond to the total magnetization and to Mn and V atoms, respectively. For both structures, the magnetic moments of the Mn atoms are multiplied by two because of the multiplicity of the position. Calculations were performed with LSDA.

Here,  $J_\nu$  is the current operator in direction  $\nu$  and  $V$  is the volume of the primitive cell.  $z^\pm$  is defined as  $E \pm i\delta$ , where  $\delta$  was chosen as 2 mRy in this study. The parameter  $\delta$  acts similarly to impurity scattering independent of spin disorder at  $T > 0$ . This value also gives rise to a residual resistivity as in previous works [54,55]. For the cases of  $L2_1$ ,  $\delta$  was introduced to prevent divergence of conductivity because resistivity cannot be defined in perfectly ordered alloys. It is noted that we restricted the range of the trace with respect to the spin indices to half of spin space to separate up and down spin components.  $\mu$  denotes the chemical potential of the system. The chemical

TABLE I. Calculated Curie temperatures, total magnetic moments, and magnetic moment of each atomic site of  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  at  $T = 0$ . Magnetic moments were calculated using  $\delta = 2$  mRy.

| System                        | $T_c^{\text{LSDA}}$ [K] | $T_c^{\text{GGA}}$ [K] | $M_{\text{Total}}^{\text{LSDA}}$ [ $\mu_B$ ] | $M_{\text{Mn}}^{\text{LSDA}}$ [ $\mu_B$ ] | $M_{\text{V}}^{\text{LSDA}}$ [ $\mu_B$ ] | $M_{\text{Total}}^{\text{GGA}}$ [ $\mu_B$ ] | $M_{\text{Mn}}^{\text{GGA}}$ [ $\mu_B$ ] | $M_{\text{V}}^{\text{GGA}}$ [ $\mu_B$ ] |
|-------------------------------|-------------------------|------------------------|----------------------------------------------|-------------------------------------------|------------------------------------------|---------------------------------------------|------------------------------------------|-----------------------------------------|
| $\text{Mn}_2\text{VAl } L2_1$ | 450                     | 620                    | 1.912                                        | 1.275                                     | -0.634                                   | 1.945                                       | 1.372                                    | -0.781                                  |
| $\text{Mn}_2\text{VGa } L2_1$ | 520                     | 720                    | 1.914                                        | 1.314                                     | -0.704                                   | 1.965                                       | 1.428                                    | -0.865                                  |
| $\text{Mn}_2\text{VAl } B2$   | 580                     | 780                    | 1.914                                        | 1.311                                     | -0.702                                   | 1.953                                       | 1.430                                    | -0.879                                  |
| $\text{Mn}_2\text{VGa } B2$   | 680                     | 920                    | 1.941                                        | 1.377                                     | -0.790                                   | 2.039                                       | 1.536                                    | -0.990                                  |

potential was found by using the following equation for the number of electrons:

$$N_e = -\frac{1}{\pi} \text{ImTr}_{iLs} \int_{-\infty}^{\infty} dE f(E, T, \mu) \int d\mathbf{e}_i \omega_i(T, \mathbf{e}_i) G_{ii}(z^+, \mathbf{e}_i). \quad (8)$$

In the numerical implementation, the chemical potential is obtained iteratively by adapting the electron/hole number condition described in Eq. (8). For a given trial value of  $\mu$ , Eq. (8) is evaluated over a wide energy range.  $\mu$  is then updated until convergence is achieved. To calculate Eq. (7), the vertex correction and coherent terms must be considered. Several studies summarized the details of the treatment of the Kubo-Greenwood formula and vertex correction term in the

TB-LMTO method [55,56]. Note that the effect of phonon scattering on the temperature dependence of physical quantities was not considered in this study. Therefore, the scattering sources for the spin-resolved conductivity were attributed only to spin and chemical disorder. A  $15 \times 15 \times 15k$ -point mesh was used for the DLM-CPA calculation to investigate the temperature dependence of magnetization. For the conductivity calculations,  $71 \times 71 \times 71k$  points were sampled in the full Brillouin zone. A mesh with  $35 \times 35 \times 35k$  points was used to calculate the spin polarization ratio from the DOS. In this study, the  $L2_1$  and  $B2$  structures are defined as shown in Fig. 1. The shown  $B2$  structure corresponds to the CsCl structure, wherein one site contains a Mn atom and the other contains equal ratios of V and Al.

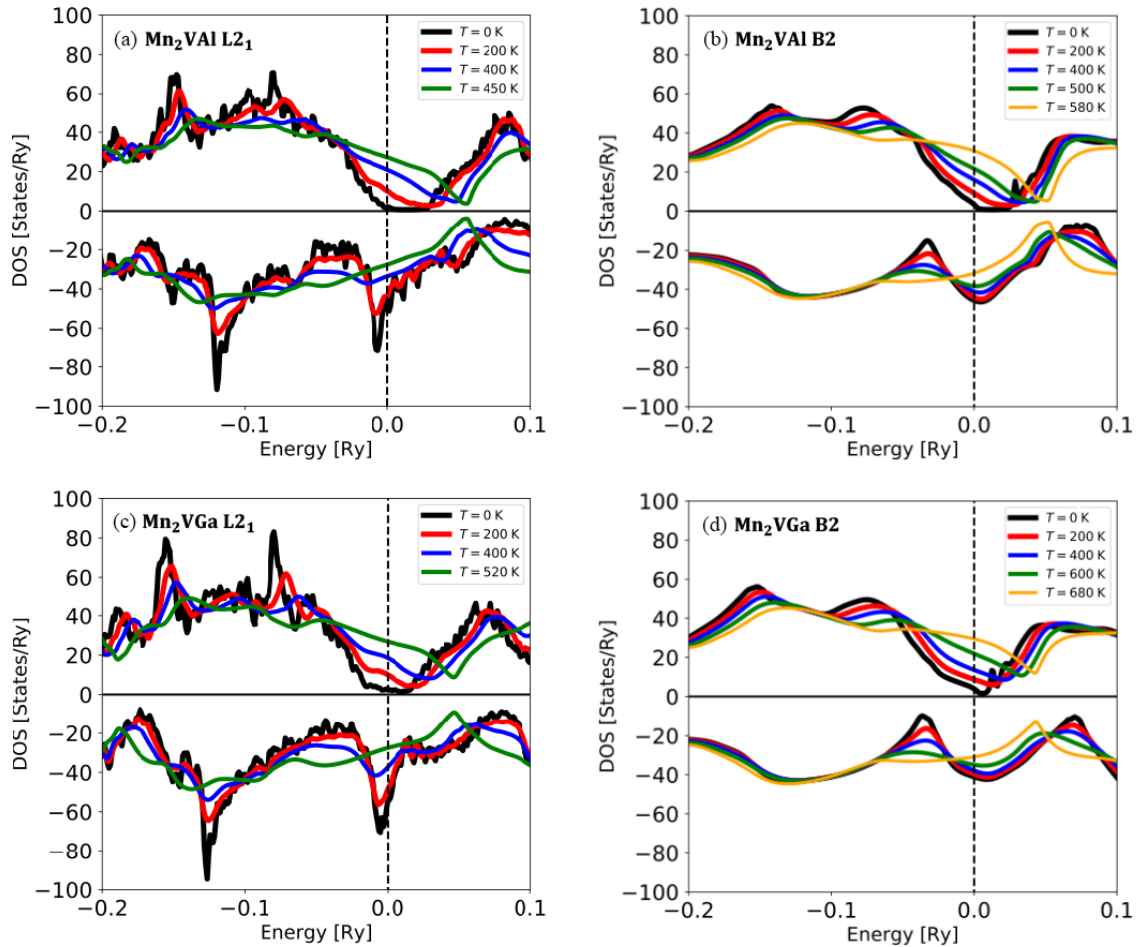


FIG. 3. Temperature dependence of the DOS of (a)  $L2_1$ - and (b)  $B2$ - $\text{Mn}_2\text{VAl}$ , and (c)  $L2_1$ - and (d)  $B2$ - $\text{Mn}_2\text{VGa}$ . Energy zero in the horizontal axis was chosen as the chemical potential at each temperature. Calculations were performed with LSDA.

### III. RESULTS AND DISCUSSION

Figure 2 shows the temperature dependence of the magnetization of  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  with  $L2_1$  or  $B2$  structure. The calculated magnetization curves resemble a Langevin function as a result of the mean-field theory of phase transition, owing to the single-site and static approximations that were used. The calculated Curie temperatures, total magnetic moments, and magnetic moment at each atomic site at zero temperature are summarized in Table I. The predicted Curie temperatures were 450 and 520 K for  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  with  $L2_1$  structure and 580 and 680 K for the alloys with  $B2$  structure. Interestingly, the predicted results indicate that the alloys with  $B2$  structure have higher Curie temperatures than those with the  $L2_1$  structure.

Experimentally, a Curie temperature of 768 K was reported for the bulk  $\text{Mn}_2\text{VAl}$  by Umetsu *et al.* [57]. For  $\text{Mn}_2\text{VGa}$ , Kumar *et al.* reported a Curie temperature of 783 K for bulk samples [53]. All of our DLM-CPA calculations based on the LSDA resulted in underestimated Curie temperatures compared to the experimental results regardless of the structure. For comparison, we applied the GGA to estimate the Curie temperatures of these systems. The following results were obtained: 620 and 720 K for the  $L2_1$  structure of  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$ , and 780 and 920 K for the  $B2$  structure of  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$ , respectively. The resulting Curie temperatures were still underestimated when the  $L2_1$  structures is assumed, although the GGA results in higher theoretical Curie temperatures compared with LSDA results. This result may originate from the fact that GGA increases the energy gap, as discussed by Weht *et al.* and Kübler [10,21]. The present treatment was based on the mean-field approximation: thus, it provides upper bounds for the Curie temperature [21] within the DLM-CPA model. Unlike localized spin Hamiltonian approaches, the present theoretical approach considers the itinerant nature of the  $d$  electrons. However, the results were not significantly different from previous theoretical predictions. One possible reason for this result is that the proposed scheme is based on the ASA. Additionally, reports have shown that spin-phonon coupling can modify the Curie temperatures of magnetic materials (see, for example, [58,59]).

Figure 3 shows the temperature dependence of the DOS for  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  with (i)  $L2_1$  and (ii)  $B2$  structures. At zero temperature, the DOS of the  $B2$  structure is smoother than that of the  $L2_1$  structure due to the broadening caused by chemical disorder scattering. In addition, these figures demonstrate that the half-metallic features of the DOS deteriorated monotonically with increasing temperature.

Previous theoretical studies have investigated the temperature dependence of the DOS of half-metallic Heusler alloys using the dynamical mean-field theory (DMFT) [60–62] and DLM-CPA [50,63–65]. In most of these previous studies, the half-metallic character of the DOS was destroyed owing to the appearance of non-quasi-particle states near the Fermi level or the alteration of the DOS by spin disorder as the temperature increased. The same trend was observed for  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  in this study. An exception was the case of  $\text{Co}_2\text{MnAl}$  with  $L2_1$  structure, as predicted by Yamashita *et al.* [50]. Additionally, Chioncel *et al.* used GGA + DMFT to calculate the temperature dependence of the spin polarization of  $\text{Mn}_2\text{VAl}$

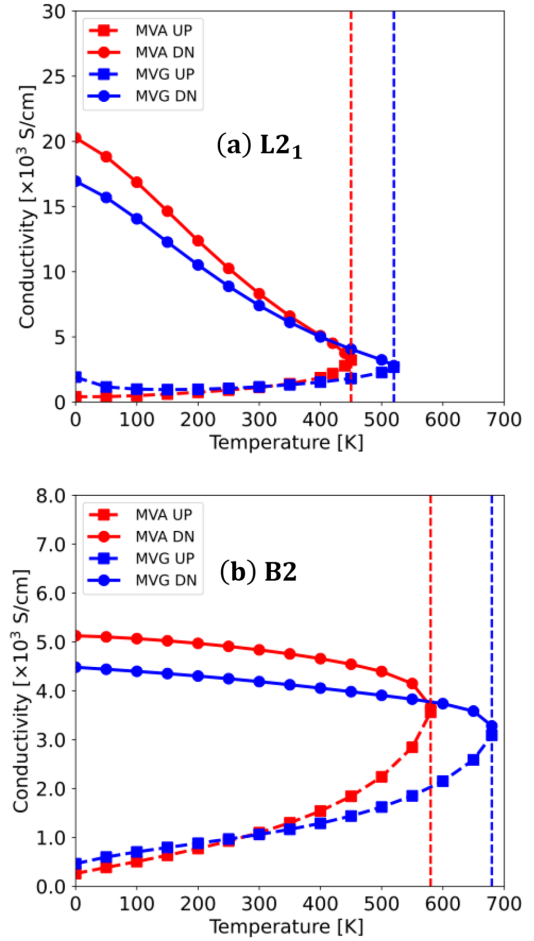


FIG. 4. Calculated temperature dependence of spin-resolved conductivities of  $\text{Mn}_2\text{VAl}$  (red) and  $\text{Mn}_2\text{VGa}$  (blue) with the (a)  $L2_1$  and (b)  $B2$  structures, respectively. Squares correspond to majority spin channel UP and circles correspond to minority spin channel DN. Vertical lines correspond to Curie temperatures for each alloy. Calculations were performed with LSDA.

from the DOS at the Fermi level [62]. Unlike DLM-CPA, their DMFT approach incorporates the dynamical components of spin fluctuations and non-quasi-particle states. However, DMFT calculations are not entirely parameter free, because a choice of the Coulomb parameter  $U$  is required. Nevertheless, their conclusions were qualitatively the same as those of the present study.

Figure 4 shows the temperature dependence of  $\sigma_{vv}^s(T)$  of  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  for both structure types. As shown in Fig. 4(a), a nearly half-metallic behavior was observed close to  $T = 0$  for both  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  with  $L2_1$  structure. This result is consistent with the calculated DOS at zero temperature. Additionally,  $\sigma_{vv}^s(T)$  of minority spin channel in the  $B2$  structure is much lower than in the  $L2_1$  structure due to chemical disorder scattering. Unlike the temperature dependence of the DOS,  $\sigma_{vv}^s(T)$  of the majority-spin channel of  $L2_1$ - $\text{Mn}_2\text{VGa}$  first decreases and then increases at low temperatures. This behavior is explained by the competition between spin-disorder scattering and changes in the density of states. The latter exhibits an increase in the number of states near the chemical potential, which closes the band gap. Both

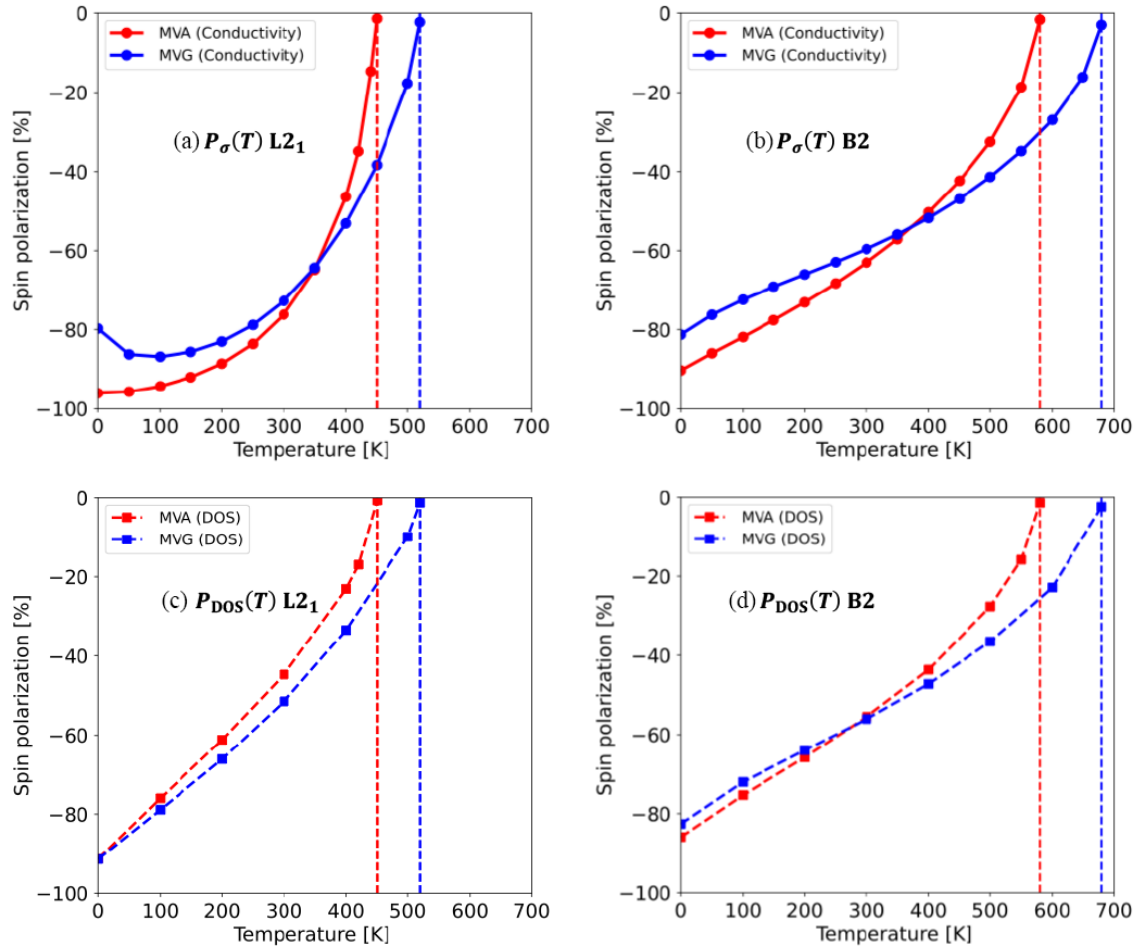


FIG. 5. Calculated temperature dependence of spin polarization calculated from (a), (b) spin resolved conductivities and (c), (d) DOS of  $\text{Mn}_2\text{VAI}$  (red) and  $\text{Mn}_2\text{VGa}$  (blue) with the  $L2_1$  and  $B2$  structures, respectively. Vertical lines correspond to Curie temperatures for each alloy. Calculations were performed with LSDA.

phenomena stem from spin transverse fluctuations at  $T > 0$ . The latter may be more significant in the case of  $L2_1$ - $\text{Mn}_2\text{VGa}$  at low temperatures and could lead to an effective improvement in spin polarization. States that appear near the chemical potential at  $T > 0$  are composed of  $d$  states, which have lower mobility than  $s$  or  $p$  states. In addition, this trend in  $\text{Mn}_2\text{VGa}$  can also be observed when  $\delta$  is chosen to be 1 mRy. The calculated residual resistivities at 0 K are  $48.36 \mu\Omega \text{ cm}$  and  $53.04 \mu\Omega \text{ cm}$  for  $\text{Mn}_2\text{VAI}$  and  $\text{Mn}_2\text{VGa}$ , respectively, when we chose  $\delta = 2$  mRy. When we chose  $\delta = 1$  mRy,  $24.80 \mu\Omega \text{ cm}$  and  $27.03 \mu\Omega \text{ cm}$  are observed for  $\text{Mn}_2\text{VAI}$  and  $\text{Mn}_2\text{VGa}$ , respectively.

These results indicate that, although the DOS and conductivity behaviors are related, they do not follow the same trends at  $T > 0$ . Therefore, the definition of half-metallicity at  $T > 0$ , whether based on DOS or conductivity, must not produce the same result. This finding is consistent with those in several previous studies [66–68]. This result can be also understood from the fact that  $\sigma_{\nu\nu}^s(T)$  depend on both the DOS and the relaxation times or velocity operator. For the  $B2$  structure, as shown in Fig. 4(b), the obtained  $\sigma_{\nu\nu}^s(T)$  of the majority spin channel increased as expected, according to the temperature dependence of the DOS. For both alloys and with both structures,  $\sigma_{\nu\nu}^s(T)$  of the minority spin channel

behaved similar to those of typical magnetic metals. It is noted that even if we choose  $\delta = 1$  mRy, the observed trend is nearly equal. The contributions from the vertex correction term were negligible for the  $L2_1$  structure. However, for the  $B2$  structure, the vertex correction term significantly affected the conductivity.

To quantify the above discussion, we present the following analysis. Once the temperature dependence of the  $\sigma_{\nu\nu}^s(T)$  is determined, the temperature dependence of the spin polarization ratio  $P_\sigma(T)$  also can be defined as follows:

$$P_\sigma(T) = \frac{\sigma_{\nu\nu}^{\text{up}}(T) - \sigma_{\nu\nu}^{\text{dn}}(T)}{\sigma_{\nu\nu}^{\text{up}}(T) + \sigma_{\nu\nu}^{\text{dn}}(T)} \times 100. \quad (9)$$

The corresponding results are presented in Fig. 5. It also shows the temperature dependence of the spin polarization ratio  $P_{\text{DOS}}(T)$ , which was evaluated from the DOS at the chemical potential, for comparison. The equation for spin polarization from the DOS is defined as

$$P_{\text{DOS}}(T) = \frac{D^{\text{up}}(T, \mu) - D^{\text{dn}}(T, \mu)}{D^{\text{up}}(T, \mu) + D^{\text{dn}}(T, \mu)} \times 100. \quad (10)$$

Here,  $\delta$  was set to 1 mRy to calculate the DOS. Reflecting the results of the temperature dependence of  $\sigma_{\nu\nu}^s(T)$ ,  $P_\sigma(T)$  of

$\text{Mn}_2\text{VGa}$  with the  $L2_1$  structure improves at low temperatures. Unlike  $P_\sigma(T)$ ,  $P_{\text{DOS}}(T)$  of  $\text{Mn}_2\text{VGa}$  decreases monotonically with increasing temperature. However, for the  $B2$  structure, both  $P_\sigma(T)$  and  $P_{\text{DOS}}(T)$  decreased monotonically with increasing temperature. In addition, the difference in the  $P_\sigma(T)$  and  $P_{\text{DOS}}(T)$  was small for the  $B2$  structure. It is noted that even if we change  $\delta$  to 1 mRy in  $L2_1$  structures, the observed behaviors of  $P_\sigma(T)$  are nearly equal.

It should be emphasized that the temperature dependence of  $P_\sigma(T)$  of  $\text{Mn}_2\text{VAl}$  with  $L2_1$  structure, was more robust than that with the  $B2$  structure. Furthermore, the difference in temperature dependence between these two spin polarizations may affect the temperature dependence of the giant and tunneling magnetoresistance differently.

It is noteworthy that the effects of phonon excitation on both  $P_\sigma(T)$  and  $P_{\text{DOS}}(T)$  can be considered within the CPA framework [69–75]. However, this aspect is beyond the scope of the present study.

Finally, the details of the energy landscape were investigated to examine whether the DLM-CPA scheme based on the force theorem is appropriate for these alloys. The total energies of the paramagnetic states were calculated self-consistently. Only the up- and down-spin states were included in the CPA calculations. The calculations were performed for  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  with the  $L2_1$  structure. In these total energy calculations, the size of the magnetic moment of one magnetic atom was varied arbitrarily using the fixed-spin-moment method while allowing the magnetic moments of the other atoms to converge freely [76]. The results are presented in Fig. 6.

Mn atoms have larger magnetic moments at zero temperature. However, the total energy landscape was shallow, as shown in Fig. 6(a). The results for the V atoms are shown in Fig. 6(b). The magnetic moment of V completely vanished in the paramagnetic state for both structures.

In addition to the LSDA method, the GGA method was used to calculate the total energy landscape of the alloys in the paramagnetic state. The results obtained were qualitatively similar to those obtained from the LSDA. These results suggest that the atoms in the alloys may exhibit significant longitudinal spin fluctuations. The energy range of thermal fluctuations of the Mn moments to cover the range of magnetic moments from 0 up to around  $2.0 \mu_B$  were approximately 700 K, which is close to the experimental Curie temperature. This property could lead to a wide range of thermal fluctuations with respect to the size of the magnetic moments. Therefore, the magnetic moments may differ from those corresponding to the energy minimum of the total energy landscape. Conversely, for V, the magnetic moments are induced and sustained by thermal fluctuations [31,77,78]. However, the results suggest that the magnetic moments of V were more itinerant than those of Mn. Nevertheless, x-ray spectroscopy experiments have shown an opposite trend.

Several authors have reported that incorporating longitudinal fluctuations can change the Curie temperatures of materials [79–81]. For example, Khemelevskiy [80] reported that longitudinal spin fluctuations can increase the critical temperature. The critical temperature of  $\text{V}_3\text{Al}$  was underestimated using a Heisenberg-type model that considered only transverse spin fluctuations. Antiferromagnetic  $\text{V}_3\text{Al}$  is

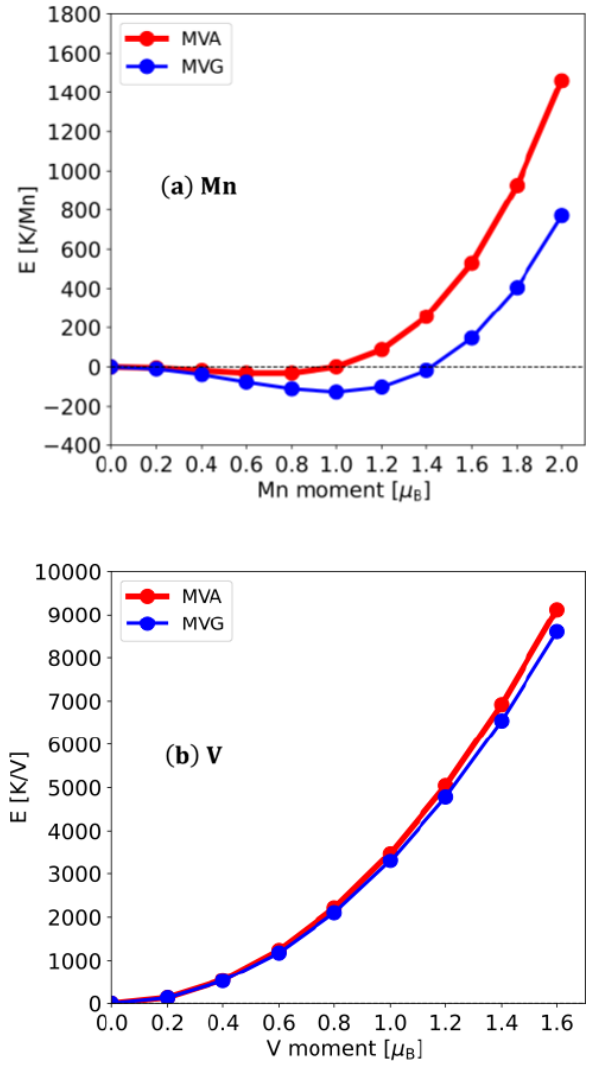


FIG. 6. Calculated total energy of paramagnetic states with different amplitude of magnetic moment of  $\text{Mn}_2\text{VAl}$  (red) and  $\text{Mn}_2\text{VGa}$  (blue) with the  $L2_1$  structures for (a) Mn and (b) V sites, respectively. Calculations were performed with LSDA.

isostructural with  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$ . The present functional integral approach, based on the force theorem, roughly corresponds to a saddle-point approximation that fixes the size of the magnetic moments at the ground-state values. However, total energy landscape calculations suggest that the most stable size of magnetic moments differs between the paramagnetic and ground states. The results also suggest the existence of a wide range of thermal fluctuations with respect to the size of the magnetic moments. These results imply that the force theorem approach may not be suitable for alloys, particularly near the Curie temperature. A treatment that reflects the energy landscape with respect to the magnetic moment amplitude can improve the theoretical results. This approach could be particularly important for predicting the Curie temperature and describing behavior near it.

#### IV. SUMMARY AND CONCLUSION

In this study, the temperature dependence of the magnetic properties, electronic structure, and spin-resolved longitudinal

transport properties of  $\text{Mn}_2\text{VAl}$  and  $\text{Mn}_2\text{VGa}$  were investigated. Calculations were performed for ordered alloys with  $L2_1$  structure and disordered alloys with  $B2$  structure. The DLM-CPA approach used here to express the transverse spin fluctuations can also incorporate both the itinerant and localized properties of the  $d$  electrons.

The predicted Curie temperatures were lower than the experimental values in most cases, which is consistent with the findings of other studies. Additionally, it was confirmed that the calculated Curie temperatures of the alloys with a  $B2$  structure were higher than those of compounds with an  $L2_1$  structure.

The calculated temperature dependence of the DOS shows a monotonic decrease in spin polarization with increasing temperature. However, in contrast to the prediction based on the behavior of the DOS at  $T > 0$ , the temperature dependence of  $\sigma_{\nu\nu}^s(T)$  of  $\text{Mn}_2\text{VGa}$  with the  $L2_1$  structure resulted in a minimum at approximately 150 K. This result may be considered a transient recovery of  $P_\sigma(T)$  around that temperature, and the minimum can be explained by the competition between the half-metal to metallic transition due to the alteration of the DOS and spin scattering. Both originate from spin disorder at  $T > 0$ . The results for the  $L2_1$  structure indicate that the DOS and conductivity behaviors do not follow the same trend at  $T > 0$ . This also means that the half-metallicity characterized by  $P_\sigma(T)$  and  $P_{\text{DOS}}(T)$  are not the same at  $T > 0$ , although they are related. On the other hand, the temperature dependences of  $P_\sigma(T)$  and  $P_{\text{DOS}}(T)$  in the  $B2$  structure are nearly the same, showing typical half-metallic behavior.

Finally, the total energy landscape in the paramagnetic states was investigated to examine the longitudinal stiffness

of the magnetic moments at different sites. The calculations were performed using the fixed-spin moment method. These results indicate that the materials exhibit strong longitudinal spin fluctuations, suggesting the itinerant nature of the  $d$  electrons. This behavior was also observed for both Mn and V magnetic moments, demonstrating that the magnetic properties of Heusler compounds and alloys cannot be simply classified as localized or itinerant.

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## DATA AVAILABILITY

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

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