

Anomalous magnetotransport behavior in Fe-doped MnNiGe alloysP. Dutta,¹ S. Pramanick,² Vijay Singh,³ Dan Thomas Major,³ D. Das,¹ and S. Chatterjee^{1,*}¹*UGC-DAE Consortium for Scientific Research, Kolkata Centre, Sector III, LB-8, Salt Lake, Kolkata 700 098, India*²*Department of Solid State Physics, Indian Association for the Cultivation of Science,**2 A & B Raja S. C. Mullick Road, Jadavpur, Kolkata 700 032, India*³*Department of Chemistry and the Lise Meitner-Minerva Center of Computational Quantum Chemistry,**Bar-Ilan University, Ramat Gan 52900, Israel*

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The electrical dc transport properties of hexagonal magnetic equiatomic alloys of nominal composition $\text{Mn}_{1-x}\text{Fe}_x\text{NiGe}$ ($x = 0.2$ and 0.25) have been investigated experimentally as well as theoretically using first-principles electronic structure calculations. Thermal hysteresis in the magnetization data indicates that the alloys undergo a first-order martensitic transition. Both the alloys show unusual nonmetallic resistivity behavior and a noticeable amount of training effect in resistivity when thermally cycled through the first-order martensitic transition. We observe moderate negative magnetoresistance ($\sim -11.5\%$ for 150 kOe) at 5 K (well below the martensitic transition temperature) associated with clear virgin line effect for both the alloys. We have adapted different flavors of density functional theory approach to understand the experimentally observed nonmetallic transport behavior.

DOI: [10.1103/PhysRevB.93.134408](https://doi.org/10.1103/PhysRevB.93.134408)**I. INTRODUCTION**

There has been a renewed interest in the transition-metal based magnetic equiatomic alloys (MEAs), primarily due to the observation of various magnetofunctional properties, such as magnetic shape memory effect, exchange bias effect (EBE), and large magnetocaloric effect (MCE) in some stoichiometric and nonstoichiometric alloys [1–7]. The stoichiometric MEA has the general formula $\text{MM}'\text{X}$, where M and M' are transition metals and X is a nonmagnetic *sp* element (Si, Ge, Sn, etc.). Among the various MEAs, MnNiGe undergoes martensitic phase transition (MPT) at 470 K during cooling and orders antiferromagnetically below 346 K [1,7]. Recently, several doping studies have been performed to enrich its functional behavior by reducing the structural transition temperature below its magnetic transition [4,5,8–10]. Coupling between magnetic and structural degrees of freedom in doped MnNiGe alloy plays a pivotal role in achieving different magneto-functional properties. Recent investigation on doped MnNiGe alloy explores different structural and magnetic aspects of this system, but no effort has been made to address their magnetotransport and hence magnetoresistance (MR) behavior. The presence of diverse magnetofunctional properties like large MCE and EBE in MEAs indicates the possibility to observe moderate MR in these alloys. Stoichiometric and off-stoichiometric Heusler based ferromagnetic shape memory alloys (FSMAs), which are the most studied materials of this kind, show reasonably large MR in the MPT region in addition to other magnetofunctional properties like large MCE, magnetostriction, and EBE [11–16]. Hence it is important to explore and understand the effect of external magnetic field on the transport properties of MnNiGe based alloys. Keeping all these in mind, the present work is focused on magnetotransport properties of Fe-doped MnNiGe alloy of nominal composition $\text{Mn}_{1-x}\text{Fe}_x\text{NiGe}$ for $x = 0.2$ and 0.25 , and this

is probably the first attempt to explore the above-mentioned properties of MnNiGe based alloys. We have also tried to explain the observed experimental data by the first-principles electronic structure calculations. Fe doping in the Mn site affects magnetic and structural transition temperatures of the sample and alters spiral antiferromagnetic (AFM) ordering by inducing the ferromagnetic (FM) or spin-glass-like state depending on the percentage of Fe doping [17]. This effect of Fe doping on the magnetic and structural properties can clearly be observed from the phase diagram developed by Liu *et al.* [17]. Observation of glassy magnetic behavior, MCE, and exchange bias tempted us to investigate these 20 and 25% Fe-doped alloys [3,6,17].

II. EXPERIMENTAL DETAILS

Polycrystalline samples of nominal composition $\text{Mn}_{1-x}\text{Fe}_x\text{NiGe}$ ($x = 0.2$ and 0.25) were prepared by argon arc melting the constituent elements of purity better than 99.9%. The ingots were then sealed in a vacuum quartz tube and annealed at 800°C for 100 h followed by rapid quenching in ice water [3,6]. Room-temperature x-ray-diffraction (XRD) patterns confirm that both the alloys are single phase with hexagonal Ni_2In -type structure [3,6]. Lattice parameters and hence the lattice volume were found to decrease linearly with increasing Fe concentration, obeying Vegard's law of alloy formation (not shown here). The resistivity (ρ) was measured using a commercial cryogen-free high-magnetic-field system from Cryogenic, Ltd., UK in the temperature (T) range 5–300 K in the presence of 0–150 kOe of external magnetic field (H). The standard four probe method was used for ρ measurements. During magnetotransport measurements, H was applied perpendicular to the direction of current. The dc magnetization (M) was measured using a Quantum Design superconducting quantum interference device magnetometer (MPMS XL 7, Evercool model).

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III. THEORETICAL TECHNIQUE

All theoretical calculations were performed on experimentally obtained unit cells using XRD and Rietveld refinement. In the Ni_2In -type austenite phase Mn and Fe are located at $2a$ (0, 0, 0) sites, Ni is at $2d$ sites ($\frac{1}{3}, \frac{2}{3}, \frac{3}{4}$), and Ge is at $2c$ sites ($\frac{1}{3}, \frac{2}{3}, \frac{1}{4}$), whereas all the atoms in the TiNiSi -type martensitic phase are located at $4c$ ($x, \frac{1}{4}, z$) sites [17]. In order to simulate the substitution of Fe at the Mn site, we constructed supercells with their size depending on the concentration (x) of Fe at the Mn site [18,19]. Specifically, to simulate 25% substitution of Fe at the Mn site, we created eight formula unit supercells (i.e., the total number of Mn in the unit cell is 8) and replaced two Mn by Fe atoms. All the density functional theory (DFT) calculations were performed using the plane-wave basis code VASP [20]. The interactions between the electrons and ions are described using the projector-augmented-wave method [20–22]. For the exchange-correlation potential, we use the generalized gradient approximation (GGA) Perdew-Burke-Ernzerhof (PBE) functional method [23]. We also used Hubbard U (on-site Coulomb interaction) values of 4.0 eV for both the Mn and Fe atoms, while the intra-atomic Hund's exchange $J = 1.0$ eV. All the aforementioned structures and cell parameters were fully relaxed with different types of magnetic structure, for instance, FM, ferrimagnetic (Ferri), and AFM ordering. The convergence of the total energy was verified with respect to the energy cutoff, which was ultimately set to 600 eV. We employed the Monkhorst-Pack scheme for k -point sampling for integration in the irreducible Brillouin zone [24]. All the calculations, including spin-orbit coupling (SOC), were performed in the noncollinear mode implemented in VASP by Hobbs *et al.* [25] and Marsman and Hafner [26]. It is important to note that, while plotting density of states (DOS) for $\text{Mn}_{0.75}\text{Fe}_{0.25}\text{NiGe}$ alloy, we consider the austenite phase with a paramagnetic (PM) ordering and the martensite phase with a FM ordering.

IV. EXPERIMENTAL RESULTS

Temperature dependences of M in the presence of 1 kOe of external H in zero-field cooled heating (ZFC), field cooling (FC), and field cooled heating (FCH) sequences for $x = 0.2$ and 0.25 alloys are depicted in Figs. 1(a) and 1(b), respectively. The zero-field cooled and field cooled conditions were achieved by cooling the sample down to 5 K from room temperature in zero field or in the presence of an applied field of 1 kOe, respectively. Clear anomalies are associated with M versus T data around 150 and 125 K for $x = 0.2$ and 0.25 alloys, respectively, indicating the presence of magnetic and martensitic transition in these alloys. Thermal hysteresis associated with FC and FCH magnetization curves confirms the first-order nature of the transition. The nature of the $M(T)$ curves observed in the studied alloys is different from the Ni-Mn-Z ($Z = \text{Sn}, \text{Sb}, \text{In}$) based Heusler-type shape memory alloys, though both show MPT [12,27,28]. In the case of Ni-Mn-Z based alloys, the high- T austenite phase has larger magnetization value than that of the low- T martensite phase [12,27,28]. On the other hand, this type of coupled magnetostructural transition from paramagnetic austenite to ferromagnetic martensite phase is also observed in some of the Heusler based

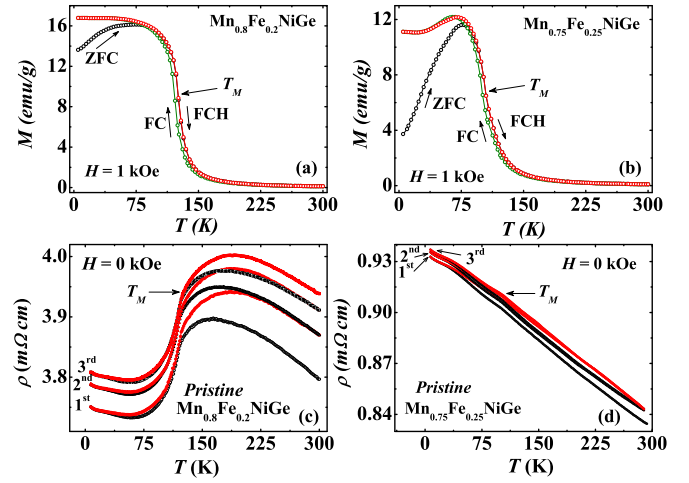


FIG. 1. (a) and (b) depict the temperature (T) variation of magnetization (M) in the presence of 1 kOe of external magnetic field (H) in zero-field cooled heating (ZFC), field cooling (FC), and field cooled heating (FCH) protocol for $x = 0.20$ and 0.25 alloys, respectively. T dependences of resistivity (ρ) data in zero magnetic field for different thermal cycles indicating the training effect are plotted in (c) and (d) for $x = 0.20$ and 0.25 alloys, respectively. Magnetostructural transition temperatures (T_M) for both the alloys are marked by arrows.

shape memory alloys (e.g., $\text{Ni}_{2+x}\text{Mn}_{1-x}\text{Ga}$, $\text{Ni}_2\text{Mn}_{1-x}\text{Cu}_x\text{Ga}$, $\text{Ni}_2\text{MnGa}_{1-x}\text{Fe}_x$, and $\text{Ni}_2\text{MnGa}_{1-x}\text{Cu}_x$) [29–32]. The FCH and ZFC data start to deviate from each other with decreasing T from just below the MPT. This indicates the development of thermomagnetic irreversibility in the system. These types of bifurcation in ZFC and FCH data are commonly observed for Heusler based FSMAs [12,27]. Martensitic variants in FSMAs act as the magnetic pinning centers and play the pivotal role towards the observed irreversibility in ZFC and FCH magnetization data [12,27].

Now let us concentrate on the thermal variation of ρ , recorded in zero field for the presently studied alloys [see Figs. 1(c) and 1(d)]. Signature of MPT is clearly visible for $x = 0.2$ alloy, whereas only a small change in slope has been observed for $x = 0.25$ alloy around the transition. Notably, both the samples show large training effect during thermal cycling through MPT in zero magnetic field. Only the first three cycles for both the samples are plotted in Figs. 1(c) and 1(d) for clear visualization. The absolute magnitude of ρ increases gradually both in the austenite and the martensite phases with thermal cycling and eventually approaches an equilibrium value after sufficient number of cycling. Thermal cycling of about 100 and 10 times saturates the training effect observed for $x = 0.2$ and 0.25 alloys, respectively. However, no noticeable change in the transition temperatures is visible within the accuracy of measurement. All further electrical measurements were performed on the trained samples (i.e., after saturating the temperature cycling effect). Apart from the training effect, there are some other interesting features also, namely, (i) nonmetallic nature of the $\rho(T)$ data below and above the MPT region ($\frac{d\rho}{dT} < 0$) and (ii) very high magnitude of ρ (~ 4000 and ~ 1000 $\mu\Omega$ cm for $x = 0.20$ and 0.25 alloys, respectively) both in the martensite and austenite phases. We

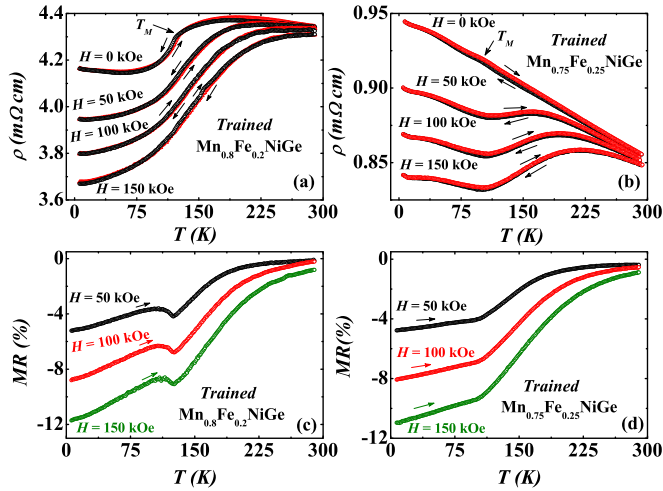


FIG. 2. (a) and (b) depict the temperature (T) variation of resistivity (ρ) for the trained samples in the presence of 0, 50, 100, and 150 kOe of applied magnetic field (H) during cooling and subsequent heating protocol for $x = 0.20$ and 0.25 alloys, respectively. Magnetotransition temperatures (T_M) for both the alloys are marked by arrows. Magnetoresistances (MR) as a function of T during heating in the presence of 50, 100, and 150 kOe of applied H for $x = 0.20$ and 0.25 alloys are plotted in (c) and (d), respectively.

have tried to analyze the $\rho(T)$ data using variable range hopping, small polaron hopping, and the Arrhenius model, but failed to explain the experimental data with these theoretical models. This indicates that the nonmetallic natures observed for the studied alloys have different origins. We have also performed some theoretical calculations (discussed later) to identify the true reason behind the observation of nonmetallic behavior.

We also recorded temperature variation of resistivity in the presence of different applied H both in cooling and heating protocols as depicted in Figs. 2(a) and 2(b). Resistivity of both the alloys is very much sensitive towards the application of external H . MPT is found to be broadened and shifted towards higher temperature in the presence of external H . On application of 150 kOe of applied H , the shift in MPT is recorded to be around 20 and 55 K for $x = 0.20$ and 0.25 alloys, respectively [shift in the MPT was determined by differentiating the $\rho(T)$ data]. Signature of MPT becomes prominent for $x = 0.25$ alloy in the presence of external H . We have also calculated and plotted the T variation of MR [MR = $\frac{\Delta\rho}{\rho_0} = \frac{\rho(H) - \rho(0)}{\rho(0)}$] at different constant applied H for both the alloys in the field cooled heating protocol [see Figs. 2(c) and 2(d)]. The alloys show negative MR (application of H results decrease in ρ) over the entire range of measurement. Magnitude of MR increases with decreasing temperature and is found to be maximum at the lowest temperature of measurement. MR is recorded to be -11.7 and -11% in the presence of 150 kOe of H around 5 K for $x = 0.20$ and 0.25 alloys, respectively. Signature of a clear dip (for $x = 0.20$) and change in slope (for $x = 0.25$) observed in the MR versus T data confirm the presence of MPT in the studied alloys.

We recorded isothermal variation of MR as a function of applied H at different constant T as depicted in Figs. 3(a) and 3(b). All the data were recorded during heating in a

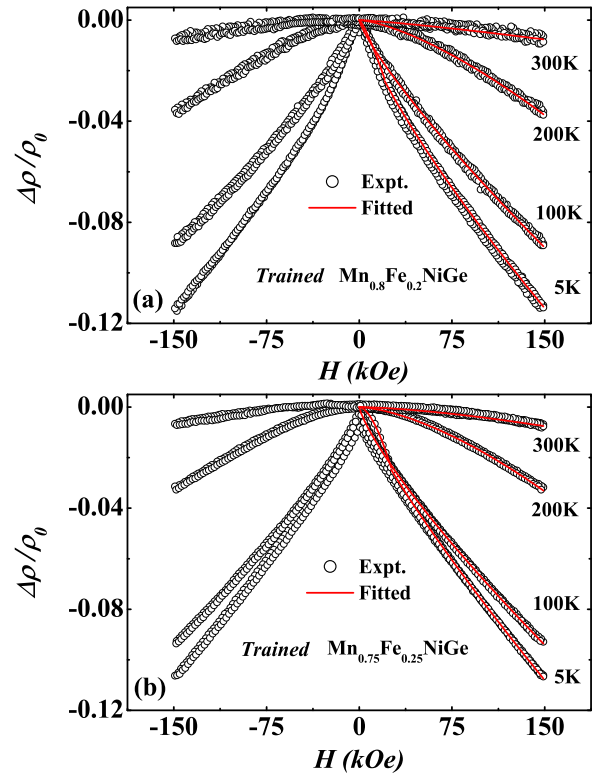


FIG. 3. Isothermal MR data are plotted as a function of applied H at different constant temperatures for (a) $x = 0.20$ and (b) 0.25 alloys. All MR vs H are recorded during heating in a thermally demagnetized state. Solid lines are the fitting to the experimental data.

thermally demagnetized state. The value of observed MR is found to be maximum at 5 K for both the alloys, which further supports the MR versus T data discussed in the previous section. No signature of field induced MPT has been observed around the MPT of any of the studied alloys. However, a clear signature of virgin line effect has been observed well below the MPT region [i.e., at 50 and 5 K, respectively (50-K data are not shown in Fig. 3 for clear visualization)] for both the alloys. To understand the true reason behind the observation of this MR behavior, magnetic-field dependence of MR is fitted with the equation $\frac{\Delta\rho}{\rho_0} = -\alpha(H)^n$ [33,34], where α is the strength of MR. The values of α and n , obtained from fitting, are summarized in Table I. The value of n at 300 K is

TABLE I. Different parameters obtained from MR vs H fitting.

x	T (K)	n	α (kOe) $^{-n}$
0.20	5	1.31	4.58×10^{-4}
	50	1.09	9.18×10^{-4}
	100	0.82	1.47×10^{-3}
	200	1.44	2.75×10^{-5}
	300	1.61	2.21×10^{-6}
0.25	5	1.78	7.10×10^{-5}
	50	0.73	2.54×10^{-3}
	100	0.80	1.70×10^{-3}
	200	1.55	1.43×10^{-5}
	300	1.94	4.60×10^{-7}

around 1.61 and 1.94 for $x = 0.20$ and 0.25 alloys, respectively. At 300 K, both the alloys should predominantly be in the PM austenite phase. Presence of short-range AFM interaction results in a deviation from the ideal value for paramagnets ($n = 2$). Sometimes a small amount of Fe doping is not enough to destroy the magnetic character (spiral AFM interaction between Mn atoms) of the parent alloy (MnNiGe in the present case), which results in the presence of some short-range AFM interaction in the system. The n is found to decrease with decreasing temperature in the PM austenite phase, whereas on further cooling below MPT it starts to increase. At 100 K, just below MPT temperature with the FM-AFM martensitic phase, observation of MR with n values of 0.82 and 0.80 for $x = 0.20$ and 0.25 alloys, respectively, may be explained on the basis of the $s - d$ scattering model, where s conduction electrons are scattered by localized d spins [33]. Well below the MPT region (at 50 and 5 K), it is evident from the figure that there are two distinct behaviors of MR versus H data at low ($H \leq 22$ kOe) and high ($H \geq 22$ kOe) field regions. The $s - d$ scattering may die out at these temperatures and consequently rules out the possibility of being responsible for the large observed MR. Magnetic and structural disorder induced localization, electron-electron interaction, and field induced AFM to FM transition are the key effects for the observation of large MR at the low- T region.

V. THEORETICAL RESULTS

We performed first-principles calculations to further dissect the effects at play in this material. We considered a variety of mechanisms previously suggested to explain nonmetallic behavior [35]. We attempted a selected subset of them in

this work to understand the experimentally observed anomaly in the resistivity data. Our analysis assumes the following: in crystalline materials the energy of electronic states is structured as bands; in a nonmetal all bands are full or empty, while in a metal one or more bands are only partially filled [36].

In the martensitic phase, the existing spiral antiferromagnetic interactions in MnNiGe are suppressed and FM correlation appears in the Mn_{0.75}Fe_{0.25}NiGe alloy, as discussed previously in this paper. In a metal, electronic states involved in charge transport (i.e., those near the Fermi energy) are spatially extended, while in a nonmetal these states are localized. The localization of the electrons in a crystalline material might be due to the following reasons: static disorder (Anderson localization); strong local electron-electron correlation, which “freezes” the local electron number (Mott transition); or strong electron-lattice coupling, which traps the electrons locally (polaronic effect). Additional mechanisms that do not involve localized states include the transition of electrons from a fully filled band (insulator) to a partially filled band (metal) under pressure or structural change. In the following, we shall attempt to model each hypothesis and analyze the results.

To understand the experimentally observed anomalous transport properties of the studied alloys, we compute the total and atom projected DOS using the PBE functional approach for the 25% Fe-doped alloy, and the results are presented in Fig. 4. Note that for convenience the partial density of states of each atom is enhanced by a factor of 2. For this calculation, we considered the following types of magnetic structures: (i) ferromagnetic interaction within and between the Mn and Fe sublattices, (ii) ferromagnetic interaction within the Mn and Fe sublattices but antiferromagnetic interaction

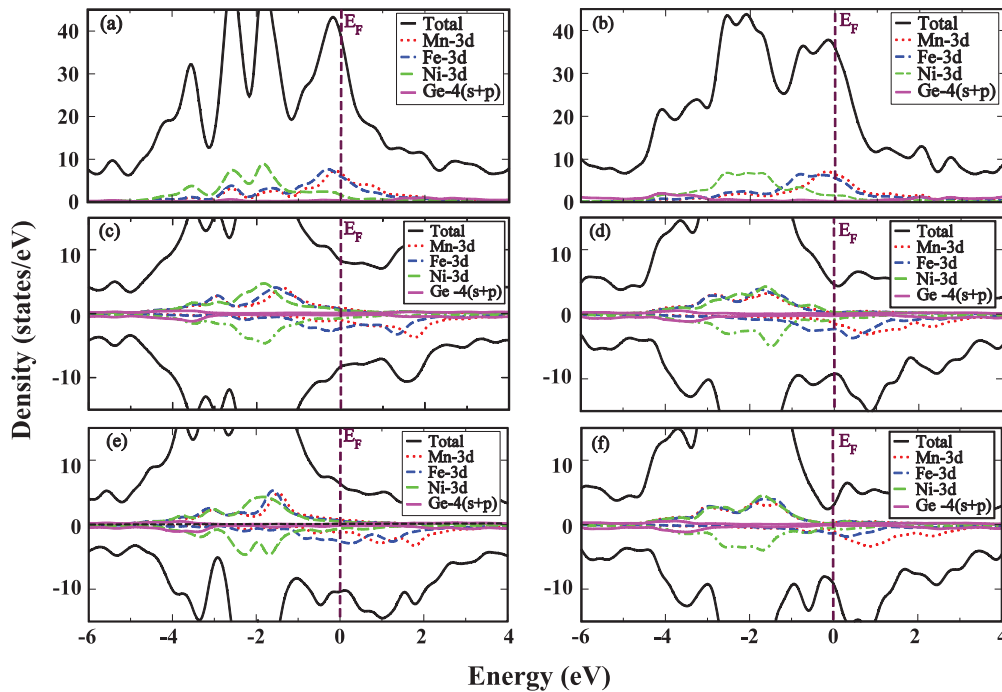


FIG. 4. Total and atom projected density of states of Mn-3d, Ni-3d, and Ge-4(s+p) for nonmagnetic (a and b), antiferromagnetic (c and d), and ferromagnetic (e and f) configurations of Mn_{0.75}Fe_{0.25}NiGe in both austenite (left panel) and martensite (right panel) phases using the PBE functional.

between the Mn and Fe sublattices, and (iii) antiferromagnetic interaction within and between Mn and Fe sublattices. Note that all subsequent calculations and analyses were performed on the ground-state magnetic structure of the system obtained using the PBE functional method. Surprisingly, we did not observe the opening of an energy gap in the DOS, and the material remains metallic in both the austenite and martensite phases. We find that the majority-spin channels of Fe-3d and Mn-3d states are completely filled. The down-spin channel of the Fe atoms is partially filled but completely empty for the Mn atoms. The above feature is present in both the austenite and martensite phases of the $\text{Mn}_{0.75}\text{Fe}_{0.25}\text{NiGe}$ alloy. Thus, the structural variation (i.e., austenite versus martensite phases) does not induce an energy gap in the DOS. Therefore, the nonmetallic nature is unlikely to be due to structural change.

Furthermore, the mechanism of the nonmetallic nature of the alloy can also be explained by either electron correlation (Mott transition) or disorder (Anderson localization), but a clear distinction is difficult [37]. First, we include the on-site Coulomb interaction ($U = 4.0$ eV) on Mn and Fe atoms, individually, to explore the possibility of a Mott insulator (i.e., band-to-localized behavior). Our computed DOS for both phases is shown in Fig. 5. We find that inclusion of Hubbard U on magnetic ions Mn [Figs. 5(a) and 5(b)] and Fe [Figs. 5(c) and 5(d)] does not open an energy gap at the Fermi level for both the austenite (left panel) and martensite (right panel) phases.

Recently Baidya and Saha-Dasgupta showed that the insulating state in $\text{La}_2\text{CoMnO}_6$ is driven by Coulomb-assisted SOC operating within the Co-d manifold [38]. We also note that recently the phenomena arising from the combined

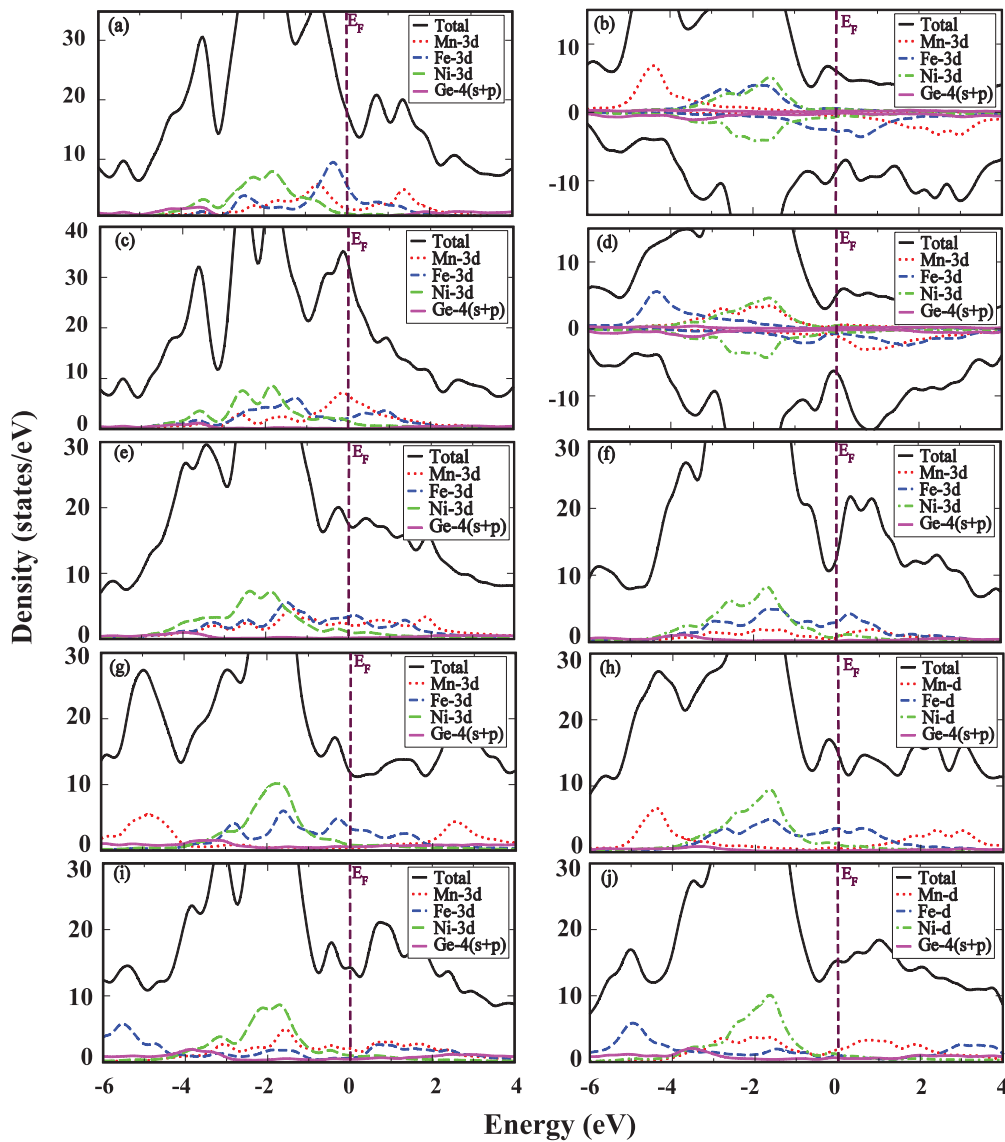


FIG. 5. Total and atom projected density of states of Mn-3d, Fe-3d, Ni-3d, and Ge-4(s+p) for both the PM austenite (left panel) and FM martensite (right panel) phases of the doped compound, $\text{Mn}_{0.75}\text{Fe}_{0.25}\text{NiGe}$, using the PBE + U functional, where $U = 4$ eV on the Mn (a and b) and Fe (c and d) atom, individually. Also shown is our computed density of states by the inclusion of SOC for the PBE functional, i.e., PBE + SOC calculation (e and f), and for the PBE + U functional, i.e., PBE + U + SOC calculation, where $U = 4$ eV on the Mn (g and h) and Fe (i and j) atom, individually.

influence of the electron correlation and SOC have been a focal point of oxides research [39]. Therefore, first, we include SOC without considering correlation in both the phases and analyze our results. Our results clearly reveal that a finite DOS exists at the Fermi level, and this is mainly composed of Fe-3d states [Figs. 5(e) and 5(f)]. Subsequently, we performed GGA + U + SOC calculations to evaluate the combined influence of electron correlation and spin-orbit coupling. The outcome was unsatisfying, even though finite orbital moments of $\sim 0.07\mu_B$ on the Fe atom and $\sim 0.02\mu_B$ on the Mn atom were observed in the FM martensite phase of the $\text{Mn}_{0.75}\text{Fe}_{0.25}\text{NiGe}$ alloy. Our results are shown in Figs. 5(g) and 5(h) for $U = 4.0$ eV on the Mn atom and in Figs. 5(i) and 5(j) for $U = 4.0$ eV on the Fe atom in both austenite and martensite phases, respectively. Again a finite DOS was obtained at the Fermi level in both the phases.

The remaining possibility of the localization may be due to static disorder (Anderson localization). Hence, we created site disorders to check disorder induced localization. We consider three cases: Mn at Ni sites (i.e., Mn@Ni), Ni at Fe sites (i.e., Ni@Fe), and Ni at Ge sites (i.e., Ni@Ge). Our results are shown in Fig. 6. We did not observe any signature of a nonmetallic state.

VI. DISCUSSIONS

We performed a systematic investigation of the electrical transport properties of two Fe-doped MnNiGe alloys of nominal compositions $\text{Mn}_{1-x}\text{Fe}_x\text{NiGe}$ ($x = 0.2$ and 0.25). Effect of temperature cycling on the transport properties is one of the key observations for the studied alloys. This type of training effect is commonly observed in the alloys having martensitic phase transition [13,40]. Large elastic strain that

develops during the nondiffusive MPT plays a pivotal role towards the observation of training effect in $\rho(T)$ data. The observed change in ρ with cycling is believed to be related to the stress-induced development of disorder (in the form of microcracks) and/or redistribution of Mn and Fe atoms causing an intrinsic change in the electronics properties [40–42]. However, no such training effect has been observed in $M(T)$ data for any of the studied alloys. Similar behavior has been observed in Ga-doped Ni-Mn-Sn and Gd_5Ge_4 alloys [13,40–42]. Apart from the temperature cycling effect, high value of ρ and its nonmetallic nature are two mentionable observations. Both nonmetallic nature and high value of resistivity data are reported for some Heusler based alloys [43]. To dissect the possible mechanism of nonmetallic nature of ρ , we performed first-principles electronic calculations using different density functional theory approaches, such as PBE, PBE + U , and PBE + U + SOC, but none of our calculations open an energy gap in the density of states. Our calculations clearly revealed that such anomalies can also not be captured by considering a static disorder in $\text{Mn}_{0.75}\text{Fe}_{0.25}\text{NiGe}$ alloy. The nonmetallic nature ($\frac{d\rho}{dT} < 0$) and high value of resistivity data in $\text{Mn}_{0.75}\text{Fe}_{0.25}\text{NiGe}$ alloy remain unclear. The nonmetallic nature observed in BaCoO_3 has also not been explained clearly by the same type of first-principles calculations using a different density functional theory approach [44]. Recent work by Kataoka tries to explain the anomalous nature of transport and magnetotransport properties of ferromagnetic metals [45]. Therefore, we propose that a temporal fluctuation (long-range Coulomb interaction), which remains one of the major limitations of DFT, should be included to explain the experimentally observed anomalies. A high-level Green's-function theory, like dynamical mean field theory, might

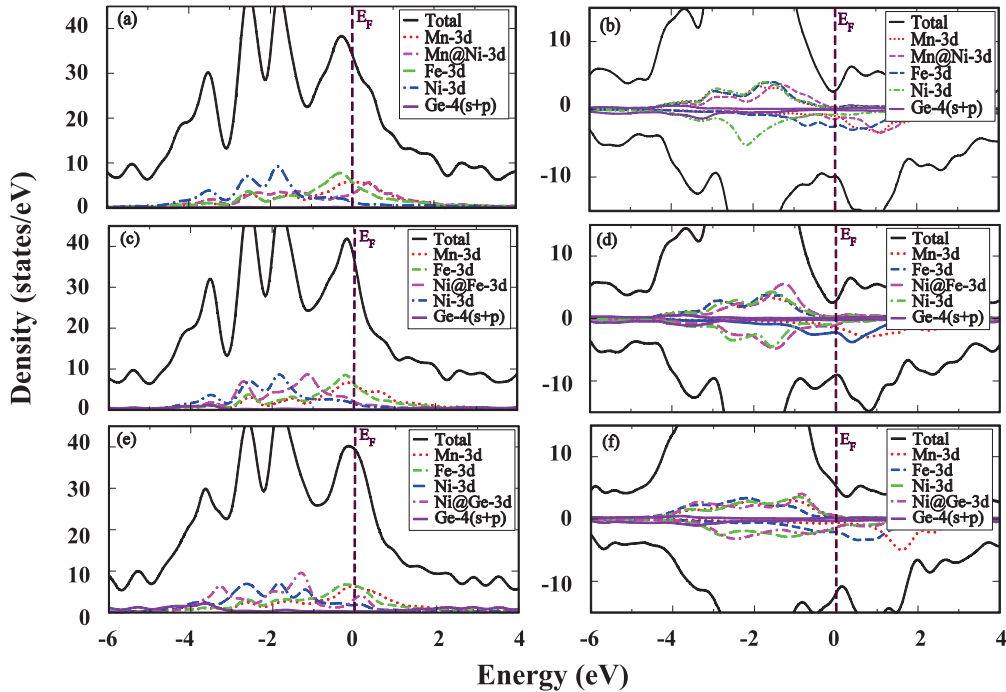


FIG. 6. Computed total and projected density of states of Mn-3d, Fe-3d, Ni-3d, and Ge-4($s+p$) for three different possible site disorders: (a, b) Mn@Ni site, (c, d) Ni@Fe site, and (e, f) Ni@Ge site in $\text{Mn}_{0.75}\text{Fe}_{0.25}\text{NiGe}$ alloy for both the austenite (left panel) and martensite (right panel) phase, respectively.

be a possible avenue to dissect the effects at play in this material [35].

Application of H induces reasonably high MR ($\sim 11\%$ for $H = 150$ kOe) in the studied alloys. Both temperature (isofield) and field (isothermal) variations of MR indicate that the low- T martensite phase is more sensitive to applied H than the high- T austenite phase. In addition, a clear virgin line effect has been observed for both the alloys well below the MPT region. The behaviors of MR (MR versus T and MR versus H) data are unlike the nature of the MR data observed for several reported metamagnetic alloys having MPT [11,14].

In conclusion, we observe a clear signature of training effect in ρ versus T data and both the alloys show reasonably high MR below the MPT region. We have tried to explain the

nonmetallic nature of $\rho(T)$ data by first-principles calculations using a different density functional theory approach. The reason behind such nonmetallic nature is not clear yet. The presence of microcracks may play an important role in this nonmetallic behavior.

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