Martensitic Transition, Ferrimagnetism and Fermi Surface Nesting in Mn_2NiGa

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Abstract

PACS. 81.30.Kf - Martensitic transformations
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The electronic structure of Mn_2NiGa has been studied using density functional theory and photoemission spectroscopy. The lower temperature tetragonal martensitic phase with c/a= 1.25 is more stable compared to the higher temperature austenitic phase. Mn_2NiGa is ferrimagnetic in both phases. The calculated valence band spectrum, the optimized lattice constants and the magnetic moments are in good agreement with experiment. The majority-spin Fermi surface (FS) expands in the martensitic phase, while the minority-spin FS shrinks. FS nesting indicates occurrence of phonon softening and modulation in the martensitic phase.

PACS numbers:

Introduction: Recent advent of multiferroic shape memory alloys (SMA) like Ni-Co-Mn-In, Ni-Mn-Ga that exhibit both ferroelastic and ferromagnetic properties has ushered a flurry of activity in this field[1,2,3,4,5,6,7,8,9]. In particular, Ni-Mn-Ga has generated immense interest because of very large strain (10%) in a moderate magnetic field (≈1 Tesla)[3,4]. Moreover, in Ni-Mn-Ga the actuation is much faster (≈2 kHz) than conventional SMA[5]. However, Ni_2MnGa are brittle and so search for materials with better mechanical properties exhibiting similar magnetic field induced strain is being actively pursued[10,11]. Mn_2NiGa is a recently discovered ferromagnetic SMA in the Ni-Mn-Ga family. It has Curie and martensitic start temperatures of 588 and 270 K, respectively[11]. Ferromagnetism in Mn_2NiGa is surprising because direct Mn-Mn interaction normally leads to antiferromagnetic alignment[12,13]. Moreover, the origin of the martensitic transition involving a relatively large tetragonal distortion (c/a= 1.21) has not been studied theoretically till date. Recently, a density functional theory (DFT) study on Mn_2NiGa shows a large enhancement of the density of states (DOS) near the Fermi level (E_F) and quenching of Mn and Ni magnetic moments in the martensitic phase[14]. However, such large change in the magnetic moments or DOS has not been observed in any other SMA either from experiment[9,15,16] or theory[8,17,18].

The geometry of the Fermi surface (FS) is responsible for a variety of phenomena like spin or charge density waves, Kohn anomalies, Friedel oscillations in metals. If the FS has parallel planes, strong electronic response can occur at the wave vector that translates one parallel plane of the FS to the other. This wave vector is called the nesting vector (n.v.). FS nesting has been reported to cause softening of the transverse-acoustic (TA_2) phonon mode along [110] direction resulting in modulated pre-martensitic phase of SMA's like Ni_2MnGa and Ni-Ti[19]. Recently, an inelastic neutron scattering study on Ni_2MnGa showed the presence of charge density wave in the martensitic phase resulting from FS nesting[7]. Thus, it is worthwhile to study the FS of Mn_2NiGa, particularly because the relatively large tetragonal distortion is likely to modify the FS substantially.

In this work, a DFT study of the electronic structure of Mn_2NiGa using full potential linearized augmented plane wave method (FPLAPW) is presented. The valence band (VB) spectrum, calculated from the theoretical DOS, is in agreement with the ultra-violet photoemission spectroscopy (UPS). We find that the total energy (E_{tot}) is lower in the martensitic phase with a tetragonal distortion of c/a= 1.25. We show that Mn_2NiGa is an itinerant ferrimagnet in both the martensitic and austenitic phases. The equilibrium lattice

constants and the magnetic moments are in agreement with x-ray diffraction and magnetization data, respectively. The FS in the martensitic phase is drastically different from the austenitic phase. A highly nested hole-type majority-spin cuboidal FS sheet around the Γ point appears in the martensitic phase that is absent in the austenitic phase.

Methodology: First principles DFT calculations were performed using the WIEN97 code[20]. Generalized gradient approximation (GGA) for the exchange correlation that accounts for the density gradients was used[21]. An energy cut-off for the plane wave expansion of 16 Ry is used ($R_{MT}K_{max}$= 9). The cut-off for charge density is G_{max}= 14. The maximum l (l_{max}) for the radial expansion is 10, and for the non-spherical part: $l_{max,ns}$=6. The muffin-tin radii are Ni: 2.1364, Mn: 2.2799, and Ga: 2.1364 a.u. The number of k points for self-consistent field cycles in the irreducible Brilloiun zone is 256 and 484 in the austenitic and martensitic phase, respectively. The convergence criterion for E_{tot} is 0.1 mRy, which implies that accuracy of E_{tot} is ±0.34 meV/atom. The charge convergence is set to 0.001. FS has been calculated using XcrySDen[22]. Mn_2NiGa ingot was prepared by arc furnace melting and annealing at 1100 K[9]. It was characterized by x-ray diffraction (XRD), energy dispersive analysis of x-rays and differential scanning calorimetry[16]. Atomically clean specimen surface was prepared by *in situ* scraping using a diamond file and the chamber base pressure was 6×10^{-11} mbar. UPS was performed with a He I (hν =21.2 eV) photon source using electron energy analyzer from Specs GmbH, Germany. The overall resolution was 120 meV.

Mn_2NiGa has a cubic L_{2_1} structure in the austenitic phase that consists of four interpenetrating f.c.c. lattices at $(0,0,0)$, $(0.25,0.25,0.25)$, $(0.5,0.5,0.5)$, and $(0.75,0.75,0.75)$ (Fig. 1a)[11,16]. The structure of Mn_2NiGa can be better explained in comparison to Ni_2MnGa that also has L_{2_1} structure. In Ni_2MnGa, the Ni atoms are at $(0.25,0.25,0.25)$ and $(0.75,0.75,0.75)$, while Mn and Ga are at $(0.5,0.5,0.5)$ and $(0,0,0)$, respectively and there is no direct Mn-Mn interaction, with Mn having eight Ni atoms as nearest neighbours. In contrast, Mn_2NiGa has one Mn atom at $(0.5,0.5,0.5)$ (referred to as MnII), while the other Mn atom (MnI) occupies the Ni atom position $(0.75,0.75,0.75)$ of Ni_2MnGa. Thus, MnI and MnII occupy inequivalent sites in the unit cell, and there is a direct Mn-Mn interaction since MnI and MnII are nearest neighbours. In the martensitic phase, the XRD pattern for Mn_2NiGa has been indexed by a tetragonal unit cell with c/a= 1.21 (Fig. 1b)[11,16].

Total energy and magnetic moment calculation: To determine whether minimization of E_{tot} causes the structural transition, we have calculated E_{tot} for both phases as a

function of the lattice parameters in the lowest energy magnetic state (discussion about the magnetic state is given later). In the austenitic phase, E_{tot} as a function of cell volume (V) exhibits a parabolic behaviour and the minimum (shown by arrow) determines the optimized lattice constant (a= 11.059 a.u.= 5.85 Å) (Fig. 2a). The agreement is within 1% of the experimental value of 5.9072 Å[11]. For the martensitic phase, in the first step, $E_{tot}(V)$ is calculated to obtain optimized V= 1330 a.u.^{3} at fixed c/a= 1.21 (XRD value). Next, $E_{tot}(c/a)$ is calculated at V= 1330 a.u.^{3}. This gives the optimized c/a to be 1.25. In the final step, $E_{tot}(V)$ is calculated again with c/a= 1.25 (Fig. 2a). Least square fitting of the data[8,23] gives the E_{tot} minimum at 1335.2 a.u.^{3} (shown by arrow). From Fig. 2a, the E_{tot} minimum in the martensitic phase is 6.8 meV/atom lower than the austenitic phase. This demonstrates that the martensitic phase is stabilized through a sizable tetragonal distortion (c/a= 1.25). The optimized lattice constants (a= 5.409 and c= 6.762, Å) are within 2.1% and 0.85% of the experimental lattice constants a= 5.5272 Å and c= 6.7044 Å, respectively[11]. Thus, the agreement of the lattice constants for both the phases is satisfying, considering that even for free-electron-like non-magnetic metals there could be about 2% discrepancy between experiment and GGA based DFT theory[24]. The decrease of V by 1.2% is in agreement with the experimental volume decrease of 0.64% in the martensitic phase[11].

The lowest energy magnetic state is obtained by performing E_{tot} minimization over various possible starting MnI and MnII magnetic moment combinations, as discussed in details in Ref.[25]. For both austenitic and martensitic phase, the anti-parallel starting spin (equal or unequal) configurations of MnI and MnII converge to a ferrimagnetic state that has minimum E_{tot}. We have used starting Mn magnetic moments for structure optimization runs to be $3\mu_B$ for both Mn atoms in anti-parallel orientation. However, when the starting MnI and MnII moments are parallel (equal or unequal), E_{tot} converges to different magnetic moments related to local minima at higher energies. For example, in the austenitic phase there are three local minima[25]. Also in the martensitic phase, multiple local minima are obtained with parallel starting moments of MnI and MnII. In particular, a local minimum that is 108 meV/atom higher in E_{tot}, gives MnI and MnII moments to be 0.24 and 2.38 μ_B[25]. Thus, one Mn moment is small, as has been reported in Ref.[14]. Our calculation based on the magnetic moments reported in Ref.[14] converges at 193 meV/atom higher energy than the E_{tot} minimum[25]. This gives an idea why the results from Ref.[14] are in disagreement with experimental data, as discussed later.

The spin magnetic moment distribution in the martensitic phase clearly shows that it is ferrimagnetic with MnI magnetic moment anti-parallel and smaller than MnII (Fig. 2b). Ni moment is small and is parallel to MnII moment. For the martensitic (austenitic) phase, the local spin magnetic moments are -2.21 (-2.43), 2.91 (3.2), 0.27 (0.32), 0.01 (0.01) μ_B per formula unit (μ_B/f.u.) for MnI, MnII, Ni, and Ga, respectively. The moment related to the interstitial charge is small (-0.04 μ_B). The total moment for the martensitic phase (1.01 μ_B/f.u.) is 11% less than the austenitc phase (1.14 μ_B/f.u). The lowering of the magnetic moment in the martensitic phase has been reported by Liu *et al.* from magnetization studies: 1.21 μ_B/f.u. (28.28 emu/g) and 1.29 μ_B/f.u. (30.3 emu/g) in the martensitic and austenitic phase, respectively[11]. Thus, the magnetic moment values and the trend that magnetization is lower in the martensitic phase are in agreement with our calculations.

Density of states and photoemission spectroscopy: The stabilization of the tetragonally distorted martensitic phase in Ni_2MnGa has been related to band Jahn-Teller effect, where a DOS peak at E_F in the cubic phase splits into two peaks below and above E_F in the tetragonal phase, resulting in a lowering of the total energy[17]. Splitting and shift of the DOS peaks just below E_F have also been observed in $Ni_{2.25}Mn_{0.75}Ga$[9]. For Mn_2NiGa, the differences in the total DOS near E_F are interesting: a peak at -0.1 eV in the austenitic phase shifts to lower energy (-0.35 eV) and diminishes in intensity in the martensitic phase (both peaks indicated by arrows). The peak above E_F at 0.35 eV (tick) does not shift but is enhanced in intensity in the martensitic phase indicating a transfer of DOS from the occupied to the unoccupied states. From the partial DOS (PDOS), it is clear that the peaks at -0.1 and -0.35 eV arise primarily due to Ni 3*d* and MnI 3*d* hybridization. The shift of the -0.1 eV peak to lower energy in the martensitic phase results from enhanced Ni 3*d*- MnI 3*d* hybridization caused by decrease in Ni-MnI distance from 2.925 Å (austenitic) to 2.701 Å (martensitic) and is a possible reason for the stabilization of the martensitic phase. The DOS at E_F is substantially reduced in the martensitic phase (1.29 states/eV f.u.) compared to the austenitic phase (3.39). Thus, decrease in electronic specific heat in the martensitic phase could be expected.

The antiferromagnetic alignment of MnI and MnII spin moments can be understood from the 3*d* spin resolved PDOS (Fig. 3b). MnI 3*d* minority-spin states appear below E_F between -1 to -3.5 eV, whereas MnII 3*d* majority-spin states appear below E_F with two well separated high PDOS region around -1.5 and -2.7 eV. MnI 3*d* majority-spin states appear primarily

above E_F centered around 0.7 eV; while MnII $3d$ minority-spin states appear above E_F with the main peak at 1.1 eV and a smaller peak at 0.35 eV. Thus, while the minority-spin states are mostly excluded from the MnII $3d$ shell, the majority-spin states are excluded from the MnI $3d$ shell resulting in large but oppositely aligned moments. MnI and MnII are nearest neighbors (n.n.) with n.n. distance of 2.549 (2.533) Å in the martensitic (austenitic) phase. The exchange pair interaction as a function of Mn-Mn separation was calculated by a Heisenberg-like model and an antiferromagnetic coupling at short interatomic distances was found that becomes ferromagnetic at larger distances[12]. Thus, direct Mn-Mn interaction at short interatomic distance is responsible for their opposite alignment[12,13]. The energy separation between the centroid of the occupied and the unoccupied spin states of opposite polarization gives an exchange splitting of 2.7 eV (3.1 eV) for MnI (MnII) in the martensitic phase. In the austenitic phase, the exchange splittings are 2.8 and 3.6 eV for MnI and MnII, respectively. Thus, the Stoner parameter (ratio of exchange splitting and magnetic moment) is roughly about 1 eV/μ_B in both phases, which is characteristic of itinerant magnetism.

It was shown for Mn excess $Ni_2Mn_{1+x}Ga_{1-x}$ that the magnetic moments of Mn atom in Ga site is equal but anti-parallel to the Mn atom at Mn site[26]. This would tend to suggest that in Mn_2NiGa, the Mn moments would cancel and a small total moment might result from Ni. However, this does not happen and the difference of MnI and MnII moments is key to the larger total moment ($\approx$1 μ_B). This originates from the stronger hybridization between the majority-spin Ni and MnII $3d$ states in comparison to hybridization between Ni and MnI $3d$ minority-spin states. Note that Ni and MnII are n.n. separated by 2.549 (2.533) Å in the martensitic (austenitic) phase and stronger hybridization pulls down almost all the MnII $3d$ majority-spin states below E_F resulting in strong spin polarization and larger moment. On the contrary, hybridization between Ni and MnI $3d$ minority-spin states is relatively weaker, distance being larger: 2.701 (2.925) Å in the martensitic (austenitic) phase, and there are sizable MnI $3d$ minority-spin states above E_F including the 0.35 eV peak, resulting in smaller moment on MnI.

Photoemission spectroscopy is a direct probe of the DOS in the VB region. In Fig. 4, the main peak of the UPS VB spectrum appears at -1.4 eV and the Fermi cut-off is at 0 eV. In order to calculate the VB spectrum, we note because of the order of magnitude larger photoemission cross-sections of Ni $3d$ and Mn $3d$ (4.0 and 5.3 mega barns at hν= 21.2 eV, respectively)[27], these PDOS determine the shape of VB[28]. So, we have added the Ni and

Mn $3d$ PDOS in proportion to their cross-sections, multiplied by the Fermi function and broadened by the instrumental Gaussian resolution and the life-time width related energy dependent Lorenzian to obtain the calculated VB (Fig. 4). This is a standard procedure of comparing the photoemission spectrum from a polycrystalline sample with the calculated DOS[28,29]. The position of the main peak at -1.4 eV and the ratio between the main peak and the intensity at E_F are in good agreement with UPS VB spectrum. It is clear from Fig. 4 that the main peak is dominated by Mn $3d$ - Ni $3d$ hybridized states that have almost equal contribution. States near E_F are dominated by Mn $3d$ states, and the MnI $3d$ in particular.

The martensitic phase DOS from Ref.[14], obtained by adding up the majority and minority-spin DOS from Fig. 5 of Ref.[14], is in clear disagreement with our DOS (Fig. 3a). This prompted us to calculate the VB spectrum from the PDOS of Ref.[14] following the same procedure as discussed above and compare it with the experimental UPS VB. As shown in Fig. 4, the calculated VB based on Ref.[14] is in obvious disagreement with UPS VB: no clear peak is observed in the former; a weak broad feature is present at -2 eV and the intensity near E_F is highest. This shows that the martensitic phase DOS reported in Ref.[14] is inconsistent with experiment. Moreover, the large change of local moments (austenitic MnI= -2.2, MnII= 3.15, Ni 0.27 μ_B to martensitic MnII= Ni $\approx$0, MnI= 1.4 μ_B) obtained in Ref.[14] is physically unexpected[25], since the MnI - MnII distance change by only 0.6% in the martensitic phase. Thus, it is no wonder why the total moment reported in Ref.[14] is higher in the martensitic phase compared to the austenitic phase, in contradiction to their own magnetization data[11,14].

Electronic bands and Fermi surface: *Austenitic phase majority spin states:* We now turn to the discussion of the electronic bands and Fermi surface of Mn_2NiGa. The majority-spin bands in the austenitic phase show that band 29 forms electron pockets (Fig. 5b). The corresponding FS, shown in Fig. 5d, is distorted prolate ellipsoidal in shape and occurs around the X point of the Brillouin zone (BZ) with the long axis along the ΓX direction. The BZ is shown in Fig. 5a. The projection of the FS along ΓX is a square (inset, Fig. 5d), which indicates that the FS nests onto itself with n.v. 0.44(1,0,0) and 0.44(0,1,0), in units of $2\pi/a$ (= 1 a.u.). The nested portion of the FS is a rhombus (shown by black lines in Fig. 5d) of area 0.052 a.u.^{2} with an opening angle of about 15°.

Martensitic phase majority spin states: In the martensitic phase, the majority-spin FS

TABLE I: Nesting vectors for the Fermi surface of Mn_2NiGa, in units of $2\pi/a$ (=1 a.u.).

	Austenitic phase		Martensitic phase	
Band no.	Majority spin	Minority spin	Majority spin	Minority spin
29	0.44(1,0,0), 0.44(0,1,0)	–	0.34(1,0,0), 0.34(0,1,0)	–
28	–	0.31{1,0,0}	0.75(1,1,0), 0.75(1,-1,0), 1.13(0,0,1)	–
27	–	0.4{1,0,0}	–	–

exhibits interesting modification (Fig. 5e). The majority-spin band 29 related electron type FS is now connected as continuous pipes along (1,0,0) direction, but with varying cross-section with flat parallel parts that nest onto each other (green/pink sheet in Fig. 5e). The n.v. are 0.34(1,0,0) and 0.34 (0,1,0), and compared to the austenitic phase the direction is same but the magnitude of the n.v.'s is reduced. Interestingly, a second majority-spin band (28) crosses E_F that results in a hole-type cuboid FS around the Γ point that has no counterpart in the austenitic phase (blue sheet, Fig. 5e). Two mutually perpendicular n.v.'s 0.75(1,1,0) and 0.75(1,-1,0) are identified, along with a larger n.v. of 1.13(0,0,1). The n.v.'s along the $\{1,0,0\}$, identified above, are not expected to contribute to phonon softening because these hardly contribute to the electron-phonon coupling matrix element[19]. On the other hand, the 0.75(1,1,0) and 0.75(1,-1,0) n.v.'s might be responsible for the softening of the TA_2[110] phonon resulting in a modulated martensitic phase. The different nesting vectors are shown in Table I.

From Fig. 5d and e, the majority spin FS is clearly enlarged in the martensitic phase compared to the austenitic phase. In the contrary, for the minority-spin states (Fig. 5f-i), the FS clearly shrinks in the martensitic phase.

Austenitic phase minority spin states: In the austenitic phase, minority spin band 27 is hole-type dispersing above (below) E_F at $0.2\Gamma L$ ($0.5LW$) and generates distorted cubic FS, where one pair of diagonally opposite corners taper out (Fig. 5f). FS nesting is observed between the cube faces with n.v. 0.4{1,0,0}, as shown by the yellow arrows. The second

sheet of the FS (band 28) is electron-like, consisting of multiply connected pipes of square cross-section (inset, Fig. 5h). The parallel surfaces of the pipes nest onto each other with a n.v. of 0.31{1,0,0} a.u. and a nesting area of 0.16 a.u.^{2}

Martensitic phase minority spin states: In the martensitic phase, the minority spin hole type FS (band 27) has a flower-like shape with a perforation in the middle (Fig. 5g). The electron type FS sheet shrink to disconnected pipes of varying diameter (Fig. 5i). These minority-spin FS sheets (Fig. 5g,i) in the martensitic phase do not exhibit nesting.

Conclusion: We observe FS nesting in the martensitic phase along [1,1,0] direction in the majority-spin FS that might lead to the instability of the TA_2 phonon mode in Mn_2NiGa. The austenitic phase FS is drastically modified in the martensitic phase. The majority spin FS expands in the martensitic phase, while the minority-spin FS shrinks. We show that Mn_2NiGa is an itinerant ferrimagnet in both austenitic and martensitic phase, and that the MnII or Ni moments do not become zero in the martensitic phase, refuting a recent work by Liu *et al.*[14]. The unequal spin magnetic moments in the two inequivalent Mn atoms (MnI and MnII) arise from the difference in the hybridization of the MnI $3d$ - Ni $3d$ and MnII $3d$ - Ni $3d$ states, which in turn is related to the interatomic distances. We furthermore show that in Mn_2NiGa a large tetragonal distortion (c/a= 1.25) decreases the total energy, stabilizing the lower temperature martensitic phase. Mn_2NiGa would be an ideal system to study different models of magnetization in metals since it has a simple L_{2_1} structure and three sublattice magnetization with parallel (between MnII and Ni) and anti-parallel (between MnI and MnII) magnetic moment alignment. Possibility of incommensurate magnetic phase or charge density wave instabilities could be expected at low temperatures due to presence of FS nesting and ferrimagnetism. Low temperature x-ray diffraction might be able to detect possible occurrence of a charge density wave state. Neutron scattering, angle resolved photoemission or Compton scattering experiments can verify the theoretically predicted FS. In fact, FS nesting, ferrimagnetism and large magnetoelastic coupling makes Mn_2NiGa a highly interesting material that has remained largely unexplored so far.

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Figure Captions

Fig. 1 The structure of Mn_2NiGa in the (a) austenitic and (b) martensitic phase; the blue, green, red, and brown spheres represent Ni, MnI, MnII and Ga, respectively.

Fig. 2 (a) The calculated total energies (E_{tot}) of Mn_2NiGa as a function of cell volume of the austenitic and martensitic phase. (b) Three dimensional plot of the spin magnetic moment distribution (in unit of $e\text{Å}^{-3}$) in the (110) plane in the martensitic phase, a contour plot is shown in the bottom.

Fig. 3 (a) Comparison of total density of states (DOS) and Ni $3d$ and Mn $3d$ partial DOS of Mn_2NiGa between the martensitic and austenitic phases (b) minority- and majority-spin components of the DOS in the martensitic phase.

Fig. 4 UPS valence band (VB) spectrum of Mn_2NiGa in the martensitic phase compared with theoretical VB spectrum calculated from the DOS in Fig. 3a. The contributions from the Mn $3d$ and the Ni $3d$ partial DOS are also shown. The spectra have been shifted along the vertical axis for clarity of presentation.

Fig. 5 (a) The f.c.c. Brillouin zone showing the high symmetry directions. (b) Majority and (c) minority-spin energy bands of Mn_2NiGa in the austenitic phase. Majority-spin Fermi surface (FS) of the (d) austenitic phase compared to the (e) martensitic phase FS related to bands 28 and 29. Minority-spin austenitic phase FS related to (f) band 27 and (h) band 28. Insets show the FS in a different orientation. Martensitic phase minority-spin FS related to (g) band 27 and (i) band 28. All the FS are shown in the repeated zone scheme and yellow arrows represent the nesting vectors. Black arrows relate the FS of the two phases.


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