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Molecular beam epitaxy growth of ferromagnetic single crystal (001) Ni_2MnGa on (001) GaAs

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The ferromagnetic shape memory alloy Ni_2MnGa has been grown on GaAs by molecular beam epitaxy. *In situ* reflection high energy electron diffraction, *ex situ* x-ray diffraction, and transmission electron microscopy selective area electron diffraction indicate the single crystal growth of a pseudomorphic tetragonal phase of Ni_2MnGa on (001) GaAs. Both vibrating sample magnetometry and superconducting quantum interference device magnetometry measurements show that the Ni_2MnGa film is ferromagnetic with in-plane magnetization and has a Curie temperature of ~320 K. © *1999 American Institute of Physics.* [S0003-6951(99)00236-3]

Microactuators have been fabricated for microelectromechanical systems (MEMS) using polycrystalline shape memory thin films.[1] As single crystal thin films are expected to have superior properties over polycrystalline ones, the epitaxial growth of single crystal films is desired,[2] but yet to be achieved. Recently, significant effort has focused on the ferromagnetic shape memory alloy Ni_2MnGa.[3–5] Bulk single crystals have shown exceptionally large magnetostriction (~4.3%).[6] These properties make thin films of Ni_2MnGa promising candidates for MEMS applications. Although a number of metallic compounds have been epitaxially grown,[7] the growth of Ni_2MnGa thin films, either polycrystalline or single crystal, has not been reported.

For stoichiometric Ni_2MnGa, the high temperature austenitic phase has the cubic Heusler ($L2_1$) crystal structure (Fig. 1) with a lattice constant of $a = 5.825$ Å. Below the martensitic transformation temperature ($T_M \sim 202$ K), the low temperature phase, martensite, is stable. It has a tetragonal crystal structure with $a = 5.92$ Å, $c = 5.57$ Å.[8] The crystal structure of the cubic phase of Ni_2MnGa can be considered as a NaCl crystal lattice of Mn and Ga with Ni occupying the tetrahedral interstitial sites. Alternatively, it may be considered as an ordered CsCl crystal structure with a simple cubic lattice of Ni atoms with every other body center site occupied by Mn and Ga, respectively. Transition metal-group-III compounds with CsCl structure and rare-earth-group-V compounds with NaCl crystal structure have been grown epitaxially on III–V semiconductors.[7] In particular, the successful epitaxial growth of NiGa (CsCl),[9] MnGa (tetragonally distorted CsCl),[10] and $Mn_{1-x}Ni_xGa$[11] on GaAs suggests that the chemically similar material Ni_2MnGa, with its 3% lattice mismatch to GaAs, may also be grown epitaxially on GaAs.

In the growth of metallic compounds on semiconductors, control of the first few atomic layers (template layer) is critical in controlling the growth orientation.[7,12] An interlayer of a different material may also act as a template layer. The close similarity of the Ni_2MnGa to the NaCl and CsCl crystal structures suggests that metallic compounds with these crystal structures may act as good templates for the epitaxial growth of Ni_2MnGa on GaAs. Here we report on the use of a lattice matched $Sc_{0.3}Er_{0.7}As$ template layer with a NaCl crystal structure.

The 0.5 μm thick GaAs buffer and 6-monolayers-thick $Sc_{0.3}Er_{0.7}As$ template layers were grown on GaAs (001) in a modified VG V80H molecular beam epitaxy (MBE) system in a similar manner to that described by Palmstrøm *et al.*[13] After the $Sc_{0.3}Er_{0.7}As$ growth the sample was allowed to cool for ~10 h facing the liquid nitrogen cooled cryopanel with a chamber pressure $<5\times10^{-11}$ mbar. The arsenic capping was performed with the sample temperature at <-10 °C using an As_4 flux. After As capping, the sample was removed from the MBE system and immediately remounted on a Mo sample holder for a RIBER-1000 MBE system, which was used for the Ni_2MnGa growth. The As cap was removed by heating the sample to ~300 °C, the resulting reflection high

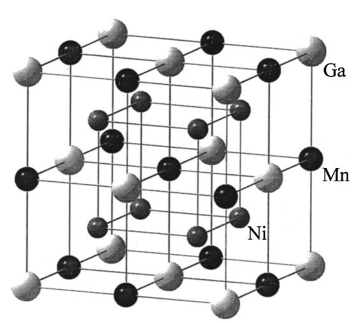

FIG. 1. Heusler $L2_1$ crystal structure of cubic Ni_2MnGa.

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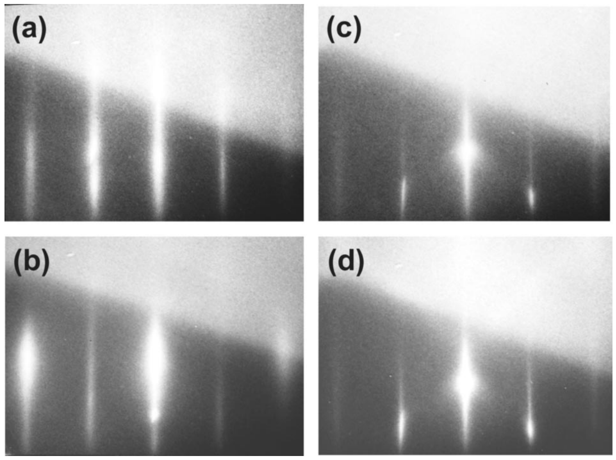

FIG. 2. RHEED patterns with the ⟨110⟩ azimuth during the epitaxial growth of Ni_2MnGa on (001) GaAs. (a) $Sc_{0.3}Er_{0.7}As$ template layer after removal of As capping at 300 °C. (b) 2× reconstructed Ni_2MnGa surface after 5 monolayers of growth at 200 °C and annealing at 300 °C. (c) Ni_2MnGa surface during codeposition at 300 °C. (d) After growth of 300 Å Ni_2MnGa at 300 °C.

energy electron diffraction (RHEED) pattern [Fig. 2(a)] from the exposed $Sc_{0.3}Er_{0.7}As$ template layer indicates a smooth unreconstructed surface. The sample was held at 300 °C for 10 min to ensure complete removal of the As cap before being cooled to 200 °C. The initial Ni_2MnGa was grown by supplying five alternate monolayers of Ni and Mn+Ga, resulting in a Ni/Mn+Ga/Ni/Mn+Ga/Ni structure. The streaky RHEED pattern obtained from this initial layer, which sharpened slightly upon annealing at 300 °C [Fig. 2(b)], corresponds to a surface unit cell of half the size of the $Sc_{0.3}Er_{0.7}As$ with a 2× reconstruction. This would be consistent with a partially ordered CsCl structure with approximately half the lattice parameter of the cubic Ni_2MnGa $L2_1$ phase with a surface reconstruction of doubled unit cell periodicity. Alternatively, it could be due to a Ni terminated surface, which would also have only half the lattice parameter of the cubic Ni_2MnGa $L2_1$ phase. Subsequent codeposition of Ni, Mn, and Ga at 300 °C corresponding to a Ni_2MnGa growth rate ~0.09 μm/h resulted in the RHEED pattern shown in Fig. 2(c). This pattern corresponds to an unreconstructed surface unit cell twice that of the one in Fig. 2(b). This is consistent with the growth of ordered Ni_2MnGa. Figure 2(d) shows the RHEED pattern after 300 Å of Ni_2MnGa growth. Kikuchi lines were clearly visible in the diffraction pattern, indicating a high quality epitaxial film. Postgrowth annealing at 300 °C for 10 min did not alter the RHEED pattern further.

Figure 3 shows a θ–2θ x-ray diffraction scan of the Ni_2MnGa/GaAs structure. Strong (002) and (004) diffraction peaks from the Ni_2MnGa thin film in addition to the (002) and (004) GaAs substrate peaks are clearly evident. This verifies the (001) Ni_2MnGa/(001) GaAs epitaxial orientation. From these data, the out-of-plane lattice constant of Ni_2MnGa was found to be 6.12 Å. Figure 4 shows selective area electron diffraction pattern from a plan-view transmission electron microscopy (TEM) specimen, which included both GaAs and Ni_2MnGa. From these data, it is clear that the Ni_2MnGa is growing pseudomorphically on GaAs with an in-plane orientation of Ni_2MnGa⟨100⟩⟨010⟩‖GaAs⟨100⟩⟨010⟩. This is a surprising result as the critical thickness for a 3% mismatch system is expected to be substantially thinner than 300 Å. Combining the x-ray diffraction and TEM selective area electron diffraction data indicates that the Ni_2MnGa is growing with a tetragonal structure with $a=b=5.65$ Å and $c=6.12$ Å on the (001) GaAs substrate. Although this phase of Ni_2MnGa has not been reported previously, we speculate that it is closely related to the orthorhombic β_1'' phase induced by uniaxial compression found by Kokorin *et al.*[14] They studied the structural transitions in bulk single crystal Ni_2MnGa as a result of ⟨110⟩ uniaxial compression and observed an orthorhombic, β_1'', phase, with $a=6.12$ Å, $b=5.78$ Å, and $c=5.54$ Å, at a uniaxial strain ~1%–2%. If this material were to be compressed biaxially along ⟨110⟩ directions, as would be the case for growth on (001) GaAs, a tetragonal phase with b and c equal to their averaged value may form. The averaged value is 5.66 Å, which is nearly identical to that found for Ni_2MnGa grown on (001) GaAs.

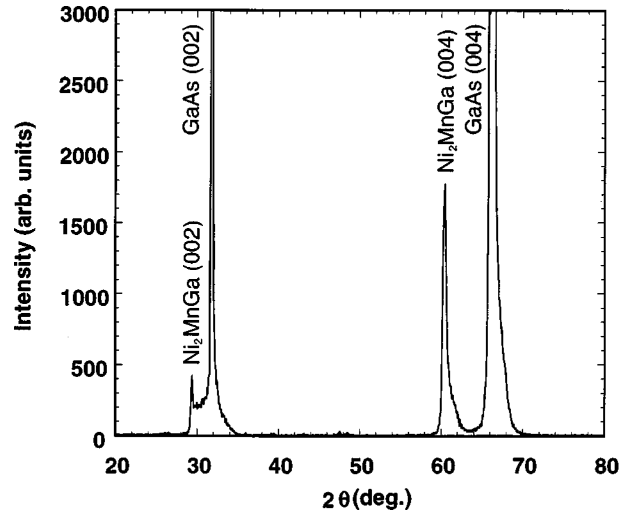

FIG. 3. X-ray diffraction scan for the 300 Å thick Ni_2MnGa film on (001) GaAs using Cu $K\alpha$ radiation.

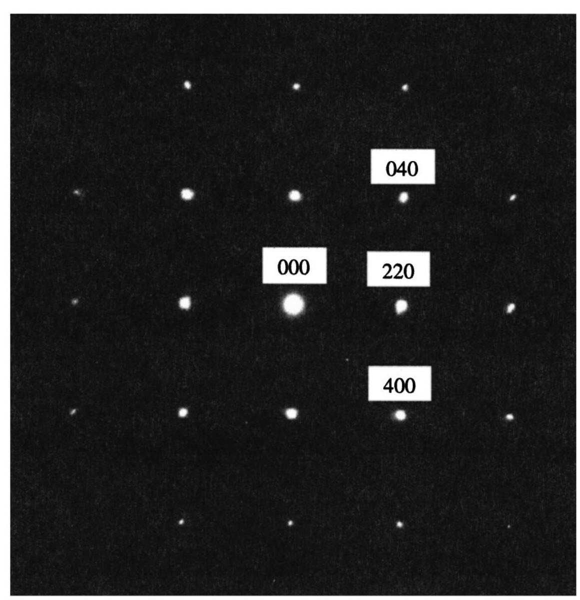

FIG. 4. Plan-view TEM selective area electron diffraction pattern along [001] zone axis. The sample is 300 Å thick Ni_2MnGa/500 Å thick GaAs.

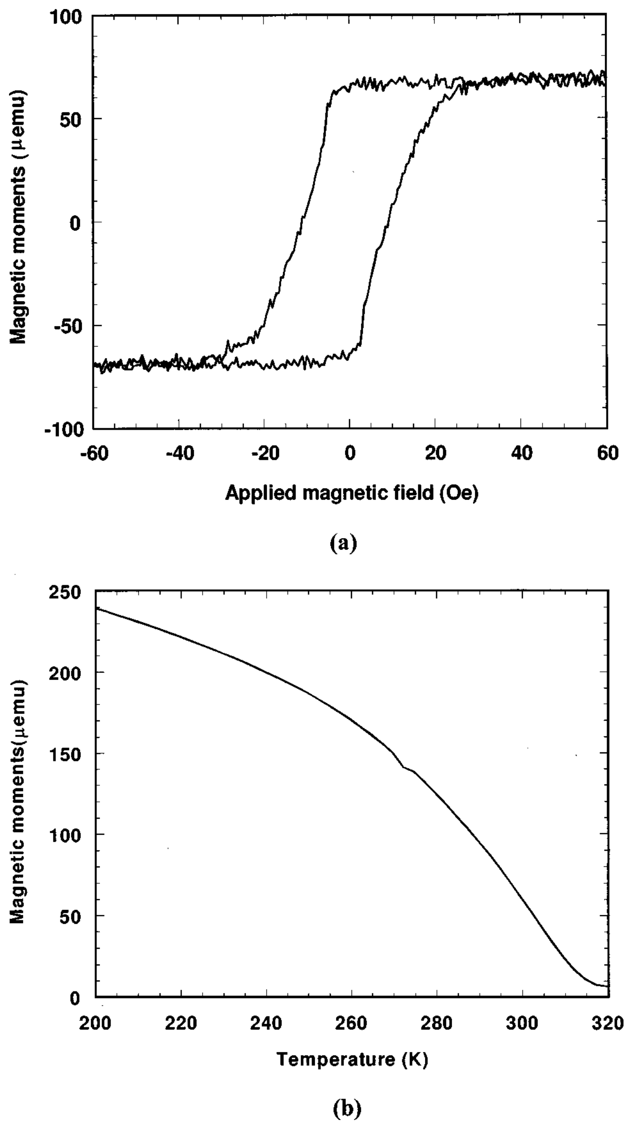

FIG. 5. Magnetic measurements for the 300 Å thick Ni_2MnGa film on (001) GaAs. (a) results from VSM measurements of in-plane magnetic moment vs magnetic field at room temperature. (b) results from SQUID measurements of the temperature dependence of in-plane magnetization.

Figure 5(a) shows the in-plane vibrating sample magnetometry (VSM) measurements for the 300 Å thick epitaxial Ni_2MnGa/GaAs sample at room temperature. The magnetic moment versus applied field curve shows a fairly square hysteresis loop with a squareness of 0.946 and a coercivity H_c of 10 Oe, both of which indicate ferromagnetic behavior. By normalizing to the film volume, the saturation magnetization was found to be ~200 emu/cm^3. The out-of-plane loop shows a large demagnetization effect and the strong influence of GaAs substrate. From our preliminary results, no strong in-plane anisotropy is observed.

Figure 5(b) shows the temperature dependence of the magnetization of the epitaxial Ni_2MnGa film obtained with superconducting quantum interference device magnetometry (SQUID) with a constant 100 Oe field applied parallel to the sample surface. These data indicate that the Curie temperature T_c is ~320 K, which explains the low coercivity of the film at room temperature. The tetragonal structure of Ni_2MnGa in this sample may be the reason for the Curie temperature being lower than that reported for the bulk stoichiometric cubic Ni_2MnGa $L2_1$ phase (~376 K).[8]

In conclusion, the growth of tetragonal ferromagnetic single crystal Ni_2MnGa ($a=b=5.65$ Å, $c=6.12$ Å) on (001) GaAs has been demonstrated. The film was pseudomorphic with the following epitaxial relationship: $(001)\langle 100\rangle\langle 010\rangle Ni_2MnGa \| (001)\langle 100\rangle\langle 010\rangle GaAs$ when grown on a $Sc_{0.3}Er_{0.7}As$ template layer.

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[1] P. Krulevitch, A. P. Lee, P. B. Ramsey, J. C. Trevino, J. Hamilton, and M. A. Northrup, J. Microelectromech. Syst. **5**, 270 (1996).
[2] R. D. James and M. Wuttig, Philos. Mag. A **77**, 1273 (1998).
[3] S. J. Murry, M. Farinelli, C. Kantner, J. K. Huang, S. M. Allen, and R. C. O'handley, J. Appl. Phys. **83**, 7297 (1998).
[4] A. N. Vasil'ev, A. D. Bozhko, V. V. Khovailo, I. E. Dikshtein, V. G. Shavrov, V. D. Buchelnikov, M. Matsumoto, S. Suzuki, T. Takagi, and J. Tani, Phys. Rev. B **59**, 1113 (1999).
[5] R. C. O'Handley, J. Appl. Phys. **83**, 3263 (1998).
[6] R. Tickle and R. D. James, J. Magn. Magn. Mater. **195**, 627 (1999).
[7] C. J. Palmstrøm, Annu. Rev. Mater. Sci. **25**, 389 (1995).
[8] P. J. Webster, K. R. A. Ziebeck, S. L. Town, and M. S. Peak, Philos. Mag. **49**, 295 (1984).
[9] A. Guivarc'h, R. Guérin, and M. Secoué, Electron. Lett. **23**, 1004 (1987).
[10] M. Tanaka, J. P. Harbison, J. DeBoeck, T. Sands, B. Philips, and T. L. Cheeks, Appl. Phys. Lett. **62**, 1565 (1993).
[11] M. Tanaka, J. P. Harbison, T. Sands, B. A. Philips, J. DeBoeck, T. L. Cheeks, L. T. Florez, and V. G. Keramidas, Mater. Res. Soc. Symp. Proc. **313**, 507 (1993).
[12] T. Sands, J. P. Harbison, C. J. Palmstrøm, R. Ramesh, and V. G. Keramidas, Mater. Res. Soc. Symp. Proc. **221**, 271 (1991).
[13] C. J. Palmstrøm, S. Mounier, T. G. Finstad, and P. F. Miceli, Appl. Phys. Lett. **56**, 382 (1990).
[14] V. V. Kokorin, V. V. Martynov, and V. A. Chernenko, Scr. Metall. Mater. **26**, 175 (1992).