Magnetoresistance in ferromagnetic shape memory alloy NiMnFeGa

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Abstract

The magnetoresistance ($MR = [R(H) - R(0)]/R(0)$) properties in ferromagnetic shape memory alloy of NiMnFeGa ribbons and single crystals, and NiFeGa ribbons have been investigated. It is found that the NiMnFeGa melt-spun ribbon exhibited GMR effect, arising from the spin-dependent scattering from magnetic inhomogeneities consisting of antiferromagnetically coupled Mn atoms in B2 structure. In the absence of these magnetic inhomogeneities, Heusler alloys seem to show a common linear MR behavior at around $0.8T_c$, regardless of sample structures. This may be explained by the $s$–$d$ model. At low temperatures, conventional AMR behaviors due to the spin–orbital coupling are observed. This is most likely due to the diminished MR from $s$–$d$ model because of much less spin fluctuation, and is not associated with martensite phase. MR anomaly at intermediate field ($\rho_x > \rho_y$) is also observed in single crystal samples, which may be related to unique features of Heusler alloys.

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1. Introduction

Ferromagnetic materials of high spin polarization have been attracting great interest due to their potential applications in spintronic devices [1]. Among them, Heusler alloys are particularly interesting since their electronic structures show 100% spin polarization [2]. In addition, certain Heusler alloys, such as Ni$_2$MnGa and NiMnFeGa, show large reversible strains in the polarization [2]. In addition, certain Heusler alloys, such as Ni$_2$MnGa and NiMnFeGa, show large reversible strains in the presence of magnetic field [3–5]. The potential new functionality combining both magnetoresistance and magnetic shape memory behaviors motivated us to investigate the magnetotransport properties of Ni$_2$MnGa and NiMnFeGa Heusler alloys.

Previous studies have shown negative MR in several Heusler alloys. For example, NiMnGa film deposited by pulsed laser deposition (PLD) on Al$_2$O$_3$ and Si substrates showed negative MR of about 3–5% over a wide temperature range [6]. It was proposed that the main contribution to MR is due to the transport between the areas with different orientation of magnetic moments at low magnetic fields, while at high fields it is an electron scattering of spin-disordered areas. In another example, nonstoichiometric polycrystalline Cu–Al–Mn shape memory alloy also show a large negative MR of about 7% at 5 K and 1% at 300 K in a magnetic field of 5 T. The MR ratio decreases after thermal annealing. It was speculated that the MR originates from the spin-dependent scattering of conduction electrons from ferromagnetic clusters embedded in a nonmagnetic matrix. Negative MR of about 1–3% at room temperature in 80 kOe field has also been observed in NiMnSb and PtMnSb [8], and is attributed to the inelastic $s$–$d$ scattering. Our group has also reported large negative MR of 9% in Fe-doped NiMnGa melt-spun ribbons [9]. Recently, a negative MR of about 5% in 80 kOe field has been discovered in bulk Ni$_2$Mn$_{1-x}$Al$_x$Ga polycrystals at room temperature [10]. These results show complexity of the magnetotransport properties in Heusler alloys and the exact mechanism remains elusive. In this paper, the origin of a large MR in NiMnFeGa ribbon sample is discussed by comparing the MR and magnetic behaviors with those of NiMnFeGa single crystal and Ni$_2$FeGa ribbon samples.

2. Experimental

Nominal Ni$_{50}$Mn$_{19}$Fe$_8$Ga$_{25}$ and Ni$_2$Ga ribbons were prepared by melt-spinning method with the same spinning linear velocity of 25 m/s. The detailed fabrication procedure is outlined elsewhere [11]. The single crystal Ni$_{50}$Mn$_{19}$Fe$_8$Ga$_{25}$ is grown by Czochralski method in Crystalox MCGS-3 cold crucible system. A 20-mm/h growth rate and a 30-rpm rotation rate were used for the growth process. The resistance and magnetization are measured along the ribbon direction and [1 0 0] direction for single crystal samples using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS-5 S).

3. Results and discussions

Fig. 1 shows the temperature dependence of resistance for Ni$_{50}$Mn$_{19}$Fe$_8$Ga$_{25}$ ribbon, Ni$_{50}$Mn$_{19}$Fe$_8$Ga$_{25}$ single crystal and...
Ni$_2$FeGa ribbon samples upon the heating process. With the increase of temperature, three samples undergo a transition from low temperature martensite phase to high temperature austenite phase. The transformation temperatures are 150, 225, and 158 K for Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ ribbon and single crystal, Ni$_2$FeGa ribbon samples, respectively. The difference in the transformation temperature between Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ ribbon and single crystal sample is attributed to the internal stress and the chemical disorder in the ribbon sample caused by fast-cooling solidification. MR measurements for three samples were performed at 5 and 300 K, corresponding to martensite and austenite phase, respectively.

Fig. 2 shows the magnetic field dependence of MR curves for Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ ribbon, Ni$_2$FeGa ribbon and Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ single crystal samples. The current and the magnetic field are parallel to the longitudinal direction of the ribbons and along the [1 0 0] direction of the single crystal sample. At 5 K, the resistance in Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ ribbon sample decreased rapidly with the increase in the magnetic field, showing a large negative MR of $\sim -13\%$ under the effect of 50 kOe field. For Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ single crystal samples, with the same nominal chemical composition as the ribbon samples and synthesized by different method, the MR reaches only $\sim -0.8\%$ in the field of 50 kOe, which is much smaller than that of the ribbon sample. The MR curve shows two regimes, corresponding to an initially rapid decrease followed by a slowly proportional decrease as the field increases from 0 to 50 kOe. For Ni$_2$FeGa ribbon, prepared under the same conditions as NiMnFeGa ribbons, a very small MR of only $\sim -1\%$ is observed at 5 K, so the effect of the quenched-in internal stress on the large MR in NiMnFeGa ribbons can be excluded. Clearly, MR behaviors for Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ ribbon are very different from those of Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ single crystal and Ni$_2$FeGa ribbon samples, and indicate spin-dependent scattering mechanism that is responsible for giant magnetoresistance (GMR). Interestingly, identical room temperature behaviors are observed in Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ single crystal and Ni$_2$FeGa ribbon samples, suggesting structural, either chemically or magnetically, independent mechanism in cubic austenite phase. Furthermore in martensite phase at 5 K, single crystal sample shows different style of low and high field MR curves that are absent in Ni$_2$FeGa ribbon samples.

To further clarify the MR behaviors, we measured longitudinal ($H \parallel I$) and transverse ($H \perp I$) MR of three samples, as shown in Fig. 3. The isotropic MR behavior in Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ ribbon again confirms the spin-dependent scattering mechanism in this sample. Isotropic MR behaviors at 300 K are also observed in Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ single crystal and Ni$_2$FeGa ribbon. At 5 K, anisotropic magnetoresistance (AMR) are observed in these two samples, with additional complications for single crystal sample at intermediate field where $\rho_{||}$ is larger than $\rho_{\perp}$, contrasting to conventional AMR behaviors.

Since both GMR and AMR behaviors are related to magnetization, we have measured initial magnetization curves [Fig. 4(a)] and magnetic hysteresis loops at 5 K [Fig. 4(b)] for three samples, respectively. The field is applied along the [1 0 0] direction in single crystals and ribbon direction. All samples are in martensite phases at 5 K, where twinning takes place to reduce the strain. The magnetocrystalline anisotropy constant ($K_1$) is typically large in martensitic phase with easy axis along [0 0 1] direction. The
single crystal sample shows a rather typical magnetization curve, a linear increase of magnetization below 5 kOe and gradual saturation at about 10 kOe, marking the transition from anomaly to normal AMR at high field. Different from the Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ single crystal sample, the magnetization of Ni$_2$FeGa ribbon increases much faster due to the much smaller magnetocrystalline anisotropy and random grain structures. By comparing these two samples, it appears that the intermediate field anomaly at 5 K (\(\rho_{\perp} > \rho_{||}\)) can only be observed in single crystal. Exact mechanism is unclear now. We speculate that the behavior may relate to the reduction of minority spin band and carrier density in this class of materials [8], which will modify the convention transport and magnetotransport behaviors in ferromagnets.

In austenite phase at room temperature, where the samples have Heusler structure, linear MR seems to be quite common, observed also in PtMnSb [8], NiMnSb [8], and Ni$_2$Mn$_{1-x}$Ga [10], regardless whether samples are in the form of single crystal, polycrystal, and thin film. The common features in this class of materials are localized spins of magnetic ions, large splitting in spin-up and spin-down band with possible disappearance of the minority band, and high carrier density [12]. The features are preserved in different structural forms and lead to a modified s–d model [12], which produces quite linear MR behaviors when \(T < 0.8T_c\) (Fig. 8 in Ref. [12]), a condition that is satisfied in our experiments. At a very low temperature, the spin fluctuation is much reduced due to stable ferromagnetic phase, the MR due to s–d model is pretty much reduced to zero [12], resulting in conventional AMR behaviors due to spin–orbit coupling.

However, Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ ribbons seems to consist of much larger magnetic inhomogeneities that gives rise to spin-dependent scattering as in the case of magnetic granular materials [13] and multilayers [14] and pins domain wall motion. The former gives rise to isotropic and negative MR and the latter manifested in the pinning mechanism reflected in the initial magnetization curve [Fig. 4(a)] and coercivity [Fig. 4(b)], which are absent in Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ single crystal and Ni$_2$FeGa ribbon.

We believe these magnetic inhomogeneities are crystalllographically constructed by antisite Mn–Mn atoms and magnetically ordered in antiferromagnetic state. As a Heusler alloy, Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ should be structured in cubic L2$_1$ structure in which the Mn and Fe atoms preferentially occupy the 4b sites (Wyckoff notation) of the cubic cell of the Fm3m structure [15]. However, Mn and Fe atoms can easily occupy other sites to form antisite B2 structure during the rapid solidification process, similar to Ni$_2$MnGa alloy [16]. These inhomogeneities in B2 structure are metastable and are relatively easier to be destroyed by thermal treatment [16]. It has been reported that the coercive force in NiMnFeGa ribbons can be reduced to zero after annealing [9]. Similarly, the value of MR can also be greatly decreased [9].

It is also known that the magnetic structure in many ordered \(X_2\)MnZ (e.g. compounds depend on the nearest Mn–Mn distance, from antiferromagnetic coupling at small separation to ferromagnetic coupling at large separations. For example, in Ni$_2$MnAl, antiferromagnetic coupling occurs in B2 structure in as-cast ingots, whereas ferromagnetic ordering appears in L2$_1$ structure after annealing [17]. Fig. 5 compares magnetization behaviors of Ni$_{50}$Mn$_{19}$Fe$_6$Ga$_{25}$ ribbon and melted ingot samples at 5 K. Reduced saturation magnetization in the ribbon sample suggests that the existence of antiferromagnetically coupled Mn ions, which cannot be aligned even at the highest field.

These antiferromagnetic clusters have random magnetization directions in zero field, giving rise to strong spin-dependent scattering. An external magnetic field will align the magnetization direction between these clusters and the ferromagnetic matrix. Consequently, spin-dependent scattering is suppressed, resulting in negative MR [14]. It should be pointed out that spins between Mn atoms inside the clusters are still antiparallel, but point to the field direction. It is now nature to raise the question why very different MR behaviors are observed in NiMnFeGa and Ni$_2$FeGa ribbons. In Ni–Fe–Ga ribbons, an increase in both Curie temperature and the saturation magnetization is clearly observed as the excess Fe atoms occupy the normal Ni sites, indicating the decrease of Fe–Fe distance in Heusler structure would not cause antiferromagnetic coupling between Fe atoms, which is opposite to the case in NiMn-based Heusler alloys [18]. Furthermore, we have confirmed that Ni$_2$FeGa ribbon has ordered Heusler structure. The (1 1 1) superlattice reflection corresponding to the nearest-neighbor L2$_1$ ordering is observed [11]. Different from NiMnFeGa ribbons, there are no magnetic inhomogeneities in Ni$_2$FeGa ribbon sample. The lack of spin-dependent scattering centers in Ni$_2$FeGa ribbon leading to normal AMR behaviors.

4. Conclusions

In summary, we have investigated the magnetoresistance properties in NiMnFeGa ribbon and single crystal, as well as in Ni$_2$FeGa ribbon samples. It is found that the NiMnFeGa melt-spun ribbons exhibited GMR effect with a large negative MR up to −13%. The spin-dependent scattering comes from the magnetic inhomogeneities consisting of antiferromagnetically coupled Mn atoms in B2 structure. In the absence of these magnetic inhomogeneities, Heusler alloys seem to show a common linear MR
behavior at around 0.8Tc, which is typically around room temperature, regardless of sample structures. This may be explained by the s−d model in ferromagnets with localized spins, large splitting spin-up and spin-down bands, and low carrier concentration, which are unique to Heusler alloys. At low temperatures, conventional AMR behaviors due to the spin–orbital coupling are observed in NiMnFeGa single crystal and Ni2FeGa ribbon samples. This is most likely due to the diminished MR from s−d model because of much less spin fluctuation. MR anomaly at field between 2 and 11 kOe (ρ− > ρ−) is also observed in single crystal sample, which may be related to unique features of Heusler alloys.

Acknowledgment

This work is supported by National Natural Science Foundation of China (Grant no. 50971130).

References