Anisotropy of the magnetoresistance in ferromagnetic shape memory alloy Ni$_{52}$Mn$_{16.4}$Fe$_8$Ga$_{23.6}$ single crystal


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Abstract

We have studied the magnetotransport properties of the quaternary Heusler alloy Ni$_{52}$Mn$_{16.4}$Fe$_8$Ga$_{23.6}$ single crystal. The magnetoresistance (MR) behavior shows a clear anisotropy at martensite state and the origin is mainly attributed to the different preferred orientations of martensite variants in three different crystallographic directions. At austenite state, the MR ratio increases linearly with increasing magnetic field and no anisotropy has been observed.

1. Introduction

Ferromagnetic shape memory alloys (FSMAs) based on the Heusler-type Ni$_2$MnGa system have been investigated intensively due to the large magnetic-field-induced strain [1–3] and their potential applications as magnetic actuator materials. In addition, some Heusler-type alloys, such as Co$_2$MnSi and NiMnSb, have been theoretically predicted to be half-metals [4], the most important materials for the spintronics. These newly discovered features of Heusler alloys have been motivating people to investigate the transport and magnetotransport properties of FSMAs-type Heusler alloys. It has been shown that the Ni$_2$MnGa and some other Heusler alloys exhibit negative magnetoresistance (MR) effect [5,6].

Lund et al. reported that the Ni$_2$MnGe films grown on GaAs(001) substrate shows a negative MR of 1% in a magnetic field of 90 kOe at 280 K [5]. For nonstoichiometric polycrystalline Cu–Al–Mn shape memory Heusler alloy, a large negative MR of 7% at 5 K and 1% at 300 K has been observed in a magnetic field of 5 T [7]. In NiMnSb and PtMnSb, MRs at 295 K in 8 T are 1% and 2.5% [8], respectively, and are attributed to the inelastic s–d scattering. Furthermore, it was found that the films of NiMnGa deposited by pulsed laser deposition (PLD) show the values of MR as high as 5% in the field of 50 kOe at 5 K and 3% at room temperature [6]. Our group has been observed the large negative MR of 9% in Fe-doped Ni–Mn–Ga melt-spun ribbons [9]. Recently, a 5% of negative MR in 8 T in bulk Ni$_{2+x}$Mn$_{1-x}$Ga polycrystals at room temperature has been reported [10]. These large negative MR values achieved seem to be encouraging for practical application. In addition, the anisotropy is an important property for MR; however, there have been no systematical studies on it in single crystal FSMAs.

In this present work, we have carried out the investigation on the anisotropy of MR in Ni$_{52}$Mn$_{16.4}$Fe$_8$Ga$_{23.6}$ single crystal.

2. Experimental

The single crystals were grown by Czochralski method in a Crystalox MCGS-3 cold crucible system under the growth conditions of 20 mm/h pulling rate and 30 rpm rotation speed. The crystallographic orientation was determined by Laue X-ray back-reflection method. The
single crystals were cut into the size of $0.5 \times 0.5 \times 10 \text{mm}^3$ with the length direction parallel to [1 0 0], [1 1 0] and [1 1 1] directions, respectively, for anisotropy of MR and magnetic measurement. The MR was measured by standard four-point fashion with current and magnetic field along the length direction of the samples. A superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS-5S) was adopted for magnetic and MR measurements.

3. Results and discussion

Fig. 1 shows the normalized electrical resistance ($R(T)/R(299 \text{K})$) as a function of temperature for Ni$_{52}$Mn$_{16.4}$Fe$_8$Ga$_{23.6}$ single crystal in the [1 0 0] direction upon heating process. It is seen that the material exhibited a distinct resistance change started at 286 K (denoted $A_S$), and terminated at 294 K (denoted $A_T$). The abrupt change in resistance occurs as a result of the reversed martensitic transformations by heating. Thus, it becomes very convenient to actually perform MR measurements and reveal the anisotropy behaviors at various states: the martensite or austenite states as well as their coexistence (at the temperature between $A_S$ and $A_T$).

Fig. 2(a-c) shows the comparative MR curves in the [1 0 0], [1 1 0] and [1 1 1] directions at 5 K (martensite state), 290 K (coexistence of martensite and austenite state) and 300 K (austenite state), respectively. All the curves were measured by varying the field in a demagnetizing way. At martensite state in Fig. 2a, the MR in the [1 0 0] direction is difficult to change in the field lower than 3 kOe, while a large increase of 0.4% occurs between 3 and 8 kOe. Further increase of field up to 50 kOe, MR increases almost linearly with field. In the [1 1 0] direction, a sharp rising of +0.4% in MR is observed at field below 0.5 kOe and the MR is followed by a decrease with increasing field. The resistance shows a decrease of about 1% from 0.5 to 25 kOe. As in the case of [1 0 0] direction, the resistance decreases linearly with increasing field. In the [1 1 1] direction, the resistance increases with increasing field up to 1 kOe, showing a positive MR of 0.25%. The MR almost keeps constant when field increases from 1 to 4 kOe and with further increase the field to 8 kOe, there is a decrease of resistance and the MR decreases to nearly zero. As the field increases higher than 8 kOe, the behavior of MR increases and is similar to those of [1 0 0] and [1 1 0]. The MRs of martensite in different directions show different behaviors, particularly under 8 kOe and a clear anisotropy can be observed.

At austenite state, the MR behaviors in three directions are almost the same, as shown in Fig. 2(c). They show almost the same linear relationship in the whole measured field range and the MR reached 2.8% under the field of 50 kOe at room temperature.

Another striking feature also can be observed from Fig. 2b: in three directions, there are apparent sharp decreases of MRs in the low-field range from 0 to 8 kOe occurred at the coexistence state, across martensitic transformation at 290 K. The MR reaches $-0.7\%$ at 8 kOe and in higher field, the MRs are conspicuous different in different directions, showing anisotropy in MR.
Although the studied sample in the austenitic phase is single crystal, the martensitic phase is essentially a polydomain state composed of variable volume fractions of the three variants. Each of these variants has a unique orientation defined by its c-axis. The application of magnetic field at martensite state can produce any of the following processes: magnetic domain wall motion, variant nucleation, twin boundary motion and magnetization rotation [11]. Any change of these processes will greatly affect the slope of the magnetization curves and thus influence the behaviors of MRs in each direction.

Fig. 3 shows the magnetization curves at 5 K in three directions. It can be seen that in [110] and [111], the curves show typical three-stage behavior in martensite. The initial rapid increase of magnetization in [110] and [111] directions from 0 to 2.5 kOe followed by the clear change in slope from 2.5 to 8 kOe and this behavior implies that the magnetization increases initially through magnetic domain wall motion and ends eventually by rotation of magnetization at high fields. The free energy minimization during magnetization requires a reconfiguration of the twinned variants, when the geometry of the magnetic domain is altered by the application of magnetic field. (Conversely, a change in twinned variants configuration by an applied field should reconfigure the magnetic domain structure.) [12]. This interaction between the variant and magnetic domain has been observed in NiMnGa [12]. In the [100] direction, the magnetization becomes harder and linearly increases with increasing field from 0 to 5 kOe and then saturates slowly as field increases to 8 kOe. It suggests that the initial state of the sample contains smaller favorable variant volume fraction in [100] than those in the other two directions, as previously reported in the case of NiMnGa materials [11]. Correspondingly, the change of MR in the [100] direction at 5 K in the region between 0 and 8 kOe is considered to be associated with the increase of favorable variant volume fraction and the rotation of moment in each variant. Since the effect of field in [110] and [111] directions is worked on domain wall motion followed by the magnetization rotation, the change of the domain configurations, and domains with magnetization parallel and perpendicular to the current direction have different resistivities. The variants in martensite have different distributions in [110] and [111] directions due to the intrinsic crystallographic effect and extrinsic factors, such as sample preparation during unbiased cooling through martensitic transformation [13]. Application of a low field of 0.5 kOe in [110] and 4 kOe in [111] directions, respectively, can readily align the domains to a parallel configuration with respect to applied field, causing resistance to decrease substantially with the alignment of magnetization in the increasing field. Therefore, the anisotropy of low-field MR in three directions reflects magnetic domain geometries and different magnetization process, including magnetic domain wall motion and magnetic moment rotation, which is related to the different preferential orientation of variants initially established in cooling and their restriction to magnetic domains in three directions. Above 10 kOe, the magnetizations in three directions almost saturate, whereas the MRs do not saturate and increase linearly with the applied field. The insaturation of MR is considered to reflect the rotation of magnetization in the domain walls. The spins in the domain walls are aligned with increasing magnetic field, which leads to the reduction of the resistance.

The austenite phase has cubic L21 structure and no obvious anisotropy of MR has been observed. The linear dependence of MR on magnetic field is similar to that has been observed in Ni2MnGa bulk and film samples, which is attributed to the s–d scattering and spin disorder scattering [10,14], as in ordinary ferromagnets.

During the martensitic transformation process (290 K), the sample showed an additional low-field effect. At this temperature, the magnetotransport properties are governed by the coexistence of different structural phases. The alloy is inhomogeneous and it is expected that three parts contributed to MR, which involves the MR from martensite and austenite, also the MR from the spin-dependent scattering by interface. It is desirable that the spin-dependent scattering through interface enhances the low-field MR, which is mainly attributed to the structure and magnetic inhomogeneous [15].

Fig. 4 shows the MR curves of [100] and [110] directions measured by varying the field 0 → 50 kOe → 0 → −50 kOe → 0 at three given temperatures. At 290 K, apparent hysteresis can be observed in both directions, while they can be negligible at 5 and 300 K. Based on the experimental result of magnetostrain in NiMnFeGa [16], these hysteresis can be attributed to field-driven twinned variants. At the same time, the interface friction involving impeding the martensite nucleation and growth [17], may also enhance the hysteresis. The different shapes of MR curves between [100] and [110] directions originate from the different arrangement and motion of variants in the

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**Fig. 3.** Normalized magnetization curves at 5 K in [100], [110] and [111] directions.
specific orientations, other than an intrinsic magnetic anisotropy of the materials. It is worth noting that the hysteresis in the field between 0 and 50 kOe are weaker than those between 0 and −50 kOe, indicating an asymmetry twin variant or interface motion with respect to these directions.

4. Summary

In conclusion, we have observed a clear anisotropy of MR at martensite state in FSMAs Ni$_{52}$Mn$_{16.4}$Fe$_8$Ga$_{23.6}$ single crystal. Our studies indicate that this anisotropy can be ascribed to the different orientation of martensite variants in different directions. During the martensitic transformation, the main contribution to MR in the low field is considered to arise from the spin-dependent transport through interface between martensite and austenite states. At austenite state, the MR can be explained by spin-disordering scattering. The anisotropy in MR at martensite state and the linear field dependence of resistance at austenite makes them attractive for applications in magnetic sensors and magnetic measurement systems.

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References