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Electronic Structures and Physical Properties of the Ordered and Disordered Ni_2MnGa Alloy Films

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Abstract

In this study, the electronic structures and physical properties of Ni_2MnGa alloy films and their dependence on the order-disorder structural transitions were investigated. The results show that the ordered films behave nearly the same as the bulk Ni_2MnGa alloy, including the martensitic transformation at 200 K. Unexpectedly, the disordering in Ni_2MnGa alloy films does not lead to any appreciable magnetic ordering down to 4 K. An annealing of the disordered films restores the ordered structure with an almost full recovery of the magnetic and the transport properties of the ordered Ni_2MnGa alloy films. A possible explanation of the disappearance of magnetic moment in the disordered film is given by using the *ab initio* first-principles electronic-structure calculations.

Keywords : Ni_2MnGa, disordered state, electronic structure

1. Introduction

Recently, Ni_2MnGa Heusler alloy (HA) attracts an exciting interest, since it exhibits a giant ferromagnetic shape-memory effect (FSME) [1,2]. This property makes Ni2MnGa thin films promising candidates for the microelectromechanical systems [3]. The ongoing works on the fabrication of Ni_2MnGa films comprise the epitaxial films prepared by molecular-beam epitaxy (MBE) on GaAs [4-7] and the sputtered polycrystalline films on Si substrates [8,9]. All these single-crystalline Ni_2MnGa films are ferromagnetic below T_C of about 320-370 K with the magnetic properties comparable to those of bulk Ni_2MnGa. However, no martensitic transformation (MT) was observed in these single -crystalline films, but according to Ref. [4] a clear sign of MT was shown for partially released films [6,7]. It is worth noting that such a transformation has been recently seen in the polycrystalline Ni_2MnGa films [8]. In this study, the electronic structures and physical properties of Ni_2MnGa alloy films and their dependence on the order-disorder structural transitions were investigated. Two distinct substrate temperatures (150 and 720 K) were chosen to achieve structures with different structural orders.

2. Experiment

An ingot of Ni_2MnGa alloy was prepared by arc melting of high purity (99.99%) elements, and subsequently melted twice, annealed at 1300 K for 6 h and then slowly cooled. The composition of was determined with x-ray fluorescence. Ni_2MnGa films of 200-300 nm in thickness were prepared by flash evaporation onto glass and NaCl substrates simultaneously. To obtain the films with a distinct order, the

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samples was deposited onto substrates heated up to 720 K and cooled down to 150 K. Additionally, some disordered films were annealed at 680 K for 60 min.

The structures of the films were characterized by using selective-area microdiffraction of transmission electron microscopy (TEM) and by x-ray diffraction (XRD). The magnetic properties were measured using SQUID and VSM in a temperature range of 4-300 K. The resistivity measurements were made in a temperature range of 4-300 K using the standard four-point probe method.

3. Results and Discussion

The structural characterization of the films deposited onto heated substrates by TEM and XRD confirmed their crystalline $L2_1$ HA structure with a mean grain size of about 40-50 nm. TEM diffraction of the films deposited onto cooled substrates revealed only a few smeared halos, typical of an amorphous structure. The mean grain size was less than the resolution limit of TEM. The annealing of the disordered films led to a crystallization to the $L2_1$ structure with a mean grain size of about 30 nm. A well-ordered crystalline state, a disordered amorphous -like state, and a crystalline state with an intermediate order were thus found in our Ni_2MnGa films, depending on the deposition temperature and the heat treatment.

Figure 1 presents the temperature dependence of magnetization, *M(T)*, of the investigated ordered and disordered films. The upper curves display the *M(T)* for the ordered Ni_2MnGa film in the field cooling (FC) at 1 kOe and the zero-field cooling (ZFC), respectively. The inset shows an enlarged portion of the ZFC curve of ordered film, together with the low-field magnetization data of bulk Ni_2MnGa. An anomaly at MT is clearly seen on the ZFC curve at T_M = 200 K, which is about 20 K lower than T_M of the bulk sample (see inset). Nearly the same behavior is observed for the film with an intermediate order after an annealing of the amorphous film (not shown). The overall

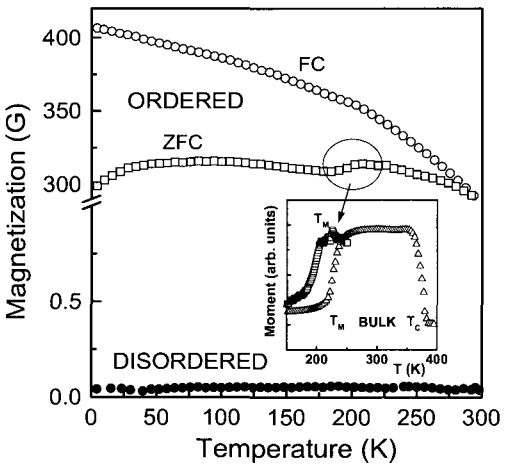

Fig. 1. In-plane M(T) curves of the ordered and disordered Ni_2MnGa films. Magnetization of the ordered film was measured in the FC cycle at 1 kOe and in the ZFC cycle. Inset shows a part of the ZFC curve, together with the low-field data of bulk sample.

magnetization behavior is nearly the same as the bulk stoichiometric Ni_2MnGa. The Curie temperature TC of the ordered film, evaluated from the VSM measurements, is 380-390 K, within an experimental error of about 10 K.

Fig. 2 shows the temperature dependence of the normalized resistance *R(T)/R(300)* for our films, together with that for the bulk alloy. The *R(T)* dependences for the films in the ordered and the intermediately-ordered states exhibit a typical metallic behavior with a positive temperature coefficient of resistivity (TCR) in the entire temperature region.

The magnetic and transport properties of disordered Ni_2MnGa films are completely different from those of the ordered films. The disordered films show a very small magnetization, nearly independent of *T*. Such a behavior could be attributed to Pauli paramagnetism. This indicates that a lack of the crystalline order in the Ni2MnGa films deposited at 150 K leads to a loss of the ferromagnetic order. The *R(T)* for the disordered film in Fig. 2 reveals a linear dependence on *T*.

In order to understand the nonmagnetic behavior of

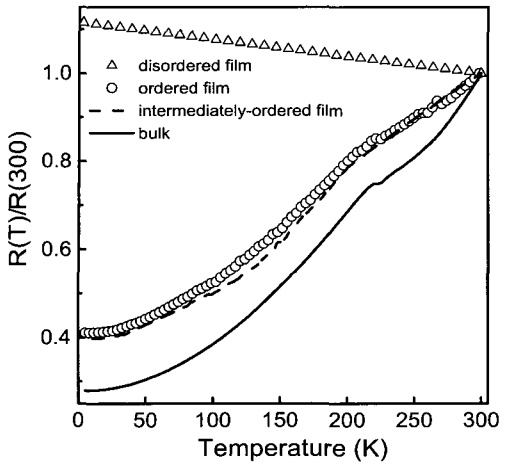

Fig. 2. Temperature dependence of the normalized resistivity of ordered, intermediately-ordered, and disordered Ni_2MnGa films. Solid line shows the same dependence of bulk Ni_2MnGa.

our disordered sample and the possible change in electron-energy spectrum upon the order-disorder transition, we carried out the electronic-structure calculation using the WIEN2k package [11].

Ni_2MnGa HA has a theoretical lattice constant of 5.811 or 5.729 Å for the ferromagnetic or the paramagnetic phase, respectively. The calculated lattice constant is very close to the experimental one. The magnetic moment turns out to be 3.982 μ_B/f.u. (3.447 μ_B/Mn and 0.292 μ_B/Ni). The crystal structure of Ni_2MnGa HA can be considered as a bcc-based fcc crystal with a chain of Ni-Mn-Ni-Ga along the (111) direction. There is a possibility of disorder in which one of the Ni atoms and the Mn atom are swapped, *i.e.*, Mn-Ni-Ni-Ga chain along the (111) direction. In this case the theoretical lattice constants come to be 5.799 and 5.719 Å for the ferromagnetic and paramagnetic phases, respectively. They are slightly smaller than the ordered case. The calculated magnetic moment is 3.261 μ_B/f.u. (2.986 μ_B/Mn), which is significantly smaller than the ordered alloy. One interesting point should be noted. The magnetic moment for Ni atom close to the Mn atom is 0.299 μ_B/Ni and almost zero for the Ni atom close to the Ga atom when the Mn-Ni-Ni-Ga chain is formed.

4. Conclusions

Ni_2MnGa HA films featuring an attractive FSME, excellent candidates for the micro/nano electro -mechanical systems and many other advanced applications, were prepared successfully by using a simple evaporation method, and the magnetic, transport and structural properties were elucidated extensively. It was also understood, for the first time, how the structural ordering in the films influences on the physical properties, including the surprising loss of ferromagnetism in the disordered films, by performing additionally the electronic-structure calculations.

Acknowledgments

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