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Investigation on the phase transition of Ni_2MnGa alloy by using impedance spectroscopy

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Abstract

The influence of structural transition on the resistance and impedance behavior of Ni_2MnGa alloy was investigated. The temperature-dependent resistance and impedance were measured in a temperature range of 4 - 350 K and 185 - 300 K, respectively. The dependence of temperature coefficient of resistivity on temperature shows a kink at 220 K, which is related to the structural transition. The change in dominant scattering mechanism results in the observed kink. Significant increases were also observed around the transition temperature for both real and imaginary parts of impedance. It is thought that this phenomenon originates from disappearance of the martensite twin boundaries during the structural transformation.

Keywords : Ni_2MnGa Heusler alloy, Impedance spectroscopy, Phase transition

1. Introduction

Ni_2MnGa, a magnetic shape-memory Heusler alloy, has recently attracted a great deal of interests due to the huge magnetorestriction for the potential applications as actuator materials. It opens a practical feasibility to control the shape and dimension of shape-memory alloy by an external magnetic field in addition to stress and temperature [1-3]. This can be realized through the reorientation of the martensitic variants under the magnetic driving force on the twin boundaries to make the magnetization of the variant parallel to the direction of the applied magnetic field, accompanying a change of dimensions. Several percent of deformation has been achieved in the Ni_2MnGa magnetic shape-memory alloys by applying a magnetic field, which is greater by an order than those of rare-earth giant-magnetostrictive alloys [4,5].

Recently, research of the Ni_2MnGa alloy is focused mainly on the magnetic properties, the magnetic-field induced shape-memory effect and the premartensitic transformation while only several investigations on the electron transport properties have been reported. Direct-current (DC) resistivity measurement was employed to study the structural and magnetic transition of NiMnGa alloy because of its simplicity and efficiency. Vasil'ev et al. observed a clear jump at the martensitic transition and a slope change at the ferro-para magnetic transition from the resistivity measurement of $Ni_{2-x}Mn_{1+x}Ga$ alloys [6]. In $Ni_{2+x}Mn_{1-x}Ga$ alloys, the resistivity measurements also show a clear jump at the martensitic transition and a discontinuous slope change at the pre-martensitic transition [7].

The DC resistivity measurements reveal an integrated contribution of the grain and the grain boundaries, which cannot be separated. Comparing to the DC measurement, both contributions are separated by the alternating-current (AC) impedance measurement. The impedance technique has been applied to widely investigate the properties of oxides [8,9]. At the same time, the giant magneto-

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impedance in amorphous materials attracted a great interest [10,11]. However, the AC characteristics of Ni_2MnGa alloy have not been studied yet, in other words, the influence of structural transition on the impedance behavior of Ni_2MnGa alloy is not clear at all. In the present work, the dependence of AC impedance characteristics on temperature is studied and compared to the DC one. The contribution of grain and grain boundaries to the impedance, and the influence of structural transition on the impedance of Ni_2MnGa alloy are discussed.

2. Experiment

The stoichiometric Ni_2MnGa alloy was prepared by melting high-purity (99.99 wt.%) Ni, Mn and Ga pieces in an arc furnace with a water-cooled Cu hearth. The subsequent homogenization of the ingot was achieved by a vacuum annealing at 1273 K for 5 h. The DC resistivity measurement was carried out in a temperature range from 4.2 to 350 K by using the four-probe method. The impedance measurement system consists of a lock-in amplifier (SR-530), an insulating box, an electrical switch, a reference resistor, a sample stage, and a computer. There are two functions of the lock-in amplifier. One is to work as an AC source and to provide a variable frequency, and the other is to measure simultaneously the voltage amplitudes of sample and reference resistor, and the phase shifts of them with respect to the reference signal. The electrical switch was used to switch the input port of lock-in amplifier between sample and reference resistor, thus the voltages at the sample and reference resistor could be measured, respectively.

3. Results and Discussion

The theory of impedance measurement can be described as follow. The voltages at reference resistor and sample can be given by

$$\tilde{U}_R \equiv V_R e^{i\theta_R} = \tilde{Z}_R I_R = r I_R \quad (1)$$

and

$$\tilde{U}_R \equiv V_s e^{i\theta_s} = \tilde{Z}_s I_s \quad (2)$$

where V is the amplitude of voltage, θ is the phase shift between voltage and reference signal, I is current, $\tilde{Z}$ is impedance, r is the resistance of reference resistor. Subscript R and S denote the reference resistor and the sample, respectively. Since the reference resistor and the sample are connected in a serial way, it is clear that

$$I_R = I_s \quad (3)$$

Combining Eqs. (1)-(3), the impedance of sample can be written as

$$\tilde{Z}_s \equiv \mathrm{Re}(\tilde{Z}_s) + i\,\mathrm{Im}(\tilde{Z}_s) = r\frac{V_s}{V_R} e^{i(\theta_S - \theta_R)}$$

where we assume the reference resistor responds linearly to current. By measuring V_s and θ_s of the sample and the reference resistor, the impedance of sample could be obtained. Calculations using a model of parallel resistor and capacitor circuit were also carried out and compared with the experiment (see below). It is found that the experiment agrees well with the calculation results. In the present work, the temperature-dependent impedance was measured from 185 to 300 K at frequencies of 50 and 100 kHz.

The dependences of DC resistance, and temperature coefficient of resistance (TCR) on temperature of Ni_2MnGa alloy are shown in Fig. 1. The resistance increases with temperature, showing a metallic behavior. On the other hand, the TCR reveals an abrupt change at a temperature of T_S (220 K). Since the resistivity reflects stringently the crystal lattice deformation, a sharp kink in the TCR curve is connected confidently with a critical temperature at which the structural transition occurs. A martensite-austenite transition from the tetragonal to cubic $L2_1$ structure was observed at 220 K for Ni_2MnGa alloy [12]. T_S of the present work coincides with the reported structural transition tempe-

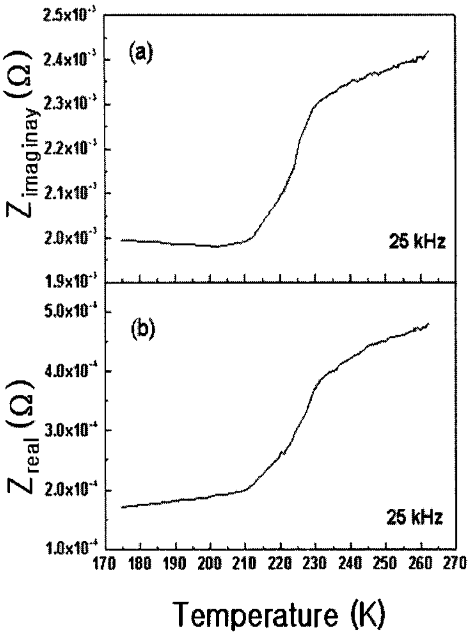

Fig. 1. Dependences of (a) DC resistance, and (b) temperature coefficient of resistance of temperature of Ni_2MnGa alloy.

rature. We analyzed the contributions of electron-magnon, electron-phonon-vibrating impurities and electron-phonon scattering on the DC resistivity of Ni_2MnGa alloy in both martensitic and austenitic states [13]. The electron-magnon and the electron-phonon scattering were found to be the dominant scattering mechanisms of Ni_2MnGa alloy in the martensitic and austenitic states, respectively. It is thought that the abrupt change around T_S in the TCR curve results from the change in dominant scattering mechanism.

The structural transition of Ni_2MnGa alloy is also studied by using the AC impedance in addition to the DC resistivity. The temperature-dependent imaginary and real parts of impedance of alloy at 25 and 100 kHz are shown in Figs. 2 and 3, respectively. A distinguished jump is observed around TS in the Figs. The real and imaginary parts of impedance reveal an abrupt increase upon the martensitic-austenitic structural transition. The imaginary part is positive, which means Ni_2MnGa alloy reveals an inductive characteristic. This is coincident

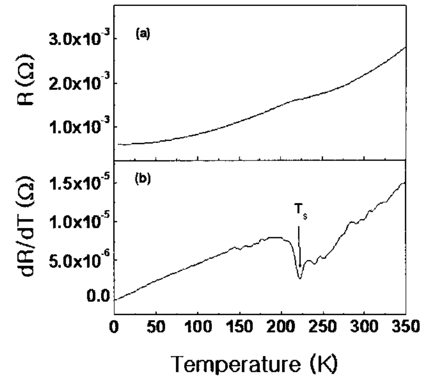

Fig. 2. Temperature-dependent (a) imaginary and (b) real parts of impedance at 25 kHz.

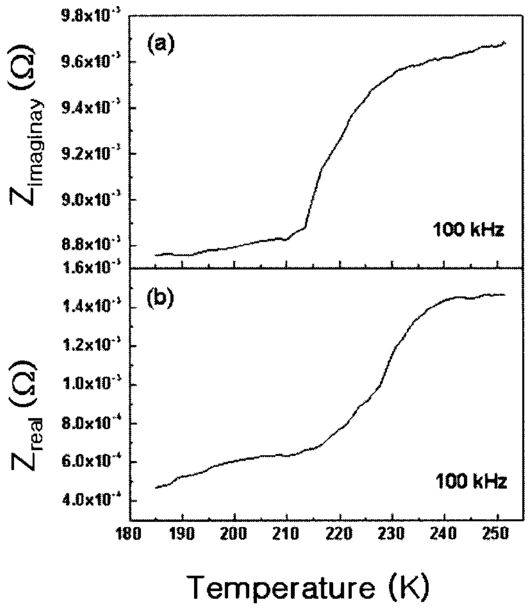

Fig. 3. Temperature-dependent (a) imaginary and (b) real parts of impedance at 100 kHz.

with the reported results for other metallic materials [14,15]. The magnitude of imaginary impedance of alloy in the austenitic state is larger than that for the martensitic state, which indicates a stronger inductive behavior in the austenitic state. To our mind, this phenomena could be interpreted as disappearance of the martensitic twin boundaries during the structural transformation.

Figure 4 is the equivalent-circuit diagram for the

AC response of Ni_2MnGa alloy. The equivalent circuit for the grains can be thought as a resistor and an inductor, which are connected in series, and that for the grain boundaries as a parallel resistor and capacitor circuit [16]. The impedance of an integral system is written by

$$\tilde{Z}-\tilde{Z}_r+i\tilde{Z}_i$$
$$=R_b+\frac{R_g}{1+\omega^2R_g^2C_g^2}+i\left(L_b-\frac{\omega R_gC_g}{1+\omega^2R_g^2C_g^2}\right) \quad (4)$$

Here, R_b and R_g are the resistances of grains and grain boundaries, respectively, L_b is the inductance produced by the grains, C_g is the capacitance of grain bound aries and interfaces, and ω is the angular frequency of current. It is known that the martensitic twins are merged by the austenitic phase, and the twin boundaries tdisappear during the martensite-austenite transition. The contribution of boundaries to the impedance is decreased correspondingly, i.e., C_g becomes smaller. According to Eq. (4), the imaginary part of impedance grows with decreasing C_g. This explains why the austenite shows a stronger inductive characteristic than the martensite. It should be noted that a significant jump during the structural transition in Figs. 2 and 3 is observed more evidently than in Fig. 2(a), which implies that the impedance technique is a more sensitive tool to elucidate the structural transition.

4. Conclusions

The influence of structural transition on the DC resistance and the AC impedance of Ni_2MnGa alloy were investigated. The temperature dependence of resistance shows a kink around the structural transition. The change in dominant scattering mechanism results the observed kink. The real and imaginary parts of AC impedance of the alloy show significant increases upon the structural transition. It is thought that disappearance of the martensite twin boundaries during the structural transformation induces the abrupt growth of the imaginary part of impedance. It is shown that the impedance technique is a more sensitive in investigating the structural transition.

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