Superconductivity of $Ni_{0.324}Nb_{0.216}Zr_{0.36}H_{0.1}$ glassy alloys†

YOSHIDA Hajime *, FUKUHARA Mikio *, INOUE Akihisa *

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1. Introduction

Glassy alloys are peculiar metallic alloys in that they lack the long-range cyclic order of crystalline alloys, on the nanoscale [1, 2]. Therefore, glassy alloys, which are considered to be macroscopic materials with a mesoscopic system of nanostructures, are candidate materials for future nanoelectronic devices. Indeed, the electronic characteristics of $(Ni_{0.36}Nb_{0.24}Zr_{0.40})_{100-y}H_y$ $(0 \le y \le 20)$ amorphous alloys are characterized by semi- superior- and superconductor and Coulomb oscillation, as hydrogen increases [3]. In the present work, we focus on superconductivity in the glassy alloys with nanoclusters, as a function of hydrogen.

Superconductivity of amorphous phases has been observed in many materials, such as, amorphous binary MoSi[4] and NbGe[5] films, and amorphous ternary $Mo_{80}P_{10}B_{10}$[6] and $Hf_{55}Nb_{30}Si_{15}$[7] ribbons. These are transition metal-metalloid-type alloys. On the other hand, superconductivity of binary and ternary metal-metal-type amorphous alloys, such as $Zr_{64}Ni_{36}$[8] and $Cu_{40}Nb_{30}Ti_{30}$[9], has also reported. However, the critical temperature for these alloys are not high, except for $Cu_{40}Nb_{30}Hf_{30}$ [9]. In the reference [3], we have reported an effect that the amount of hydrogen content gave to the superconductivity characteristic in terms of electromagnetic interaction under strong magnetic field. As far as we know, there are no previous reports on this subject for glassy alloys with hydrogen, and also no distinct interpretation for superconductivity of glassy alloys.

2. Experimental Procedure

The rotating wheel method under an argon atmosphere was used for the preparation from argon arc-melted ingots of amorphous $Ni_{36}Nb_{24}Zr_{40}$ alloy ribbons of about 1-mm width and 30-μm in thickness. Hydrogen charging was carried out electrolytically in 0.5 M H_2SO_4 and 1.4 g /L thiourea (H_2NCSNH_2) at room temperature and at the current densities of 30 A/m^2. The amounts of hydrogen absorbed in the specimens were measured by the inert gas carrier melting-thermal conductivity method. The structure of the $(Ni_{0.36}Nb_{0.24}Zr_{0.40})_{100-y}H_y$ $(0 \le y \le 0.6)$ glassy alloys was identified by X-ray diffraction with Cu Kα radiation at grazing incident mode. The specific electrical resistance of hydrogenated specimens was measured by the four-probe method DC and AC Current Source 6221, Nano Voltmeter 2182A; (Keithley Instruments Inc.) with a DC current of ±1 mA at cooling and heating rates of 1 K/s from 373 K to 5.5 K in He of ambient pressure (Top Loading Refrigerated Cryostat; JECC Torisha Co.). The distance between the two voltage electrodes was 20 mm. Electrical resistance measurements of superconductivity below 4 K were determined by the temperature dependence of the DC current four-probe method at a cooling rate of 1 K/min from 323 K to 1.4 K in He of ambient pressure under a magnetic in High Field Laboratory (IMR, Tohoku University).

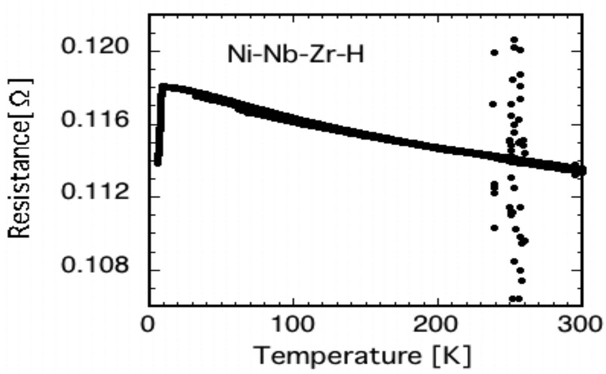

Fig. 1 Temperature dependence of electrical resistance for Ni-Nb-Zr-H glassy alloy.

3. Experimental Results

We show the temperature dependence of electrical resistance for Ni-Nb-Zr-H in **Fig. 1**. In a cooling run from 300 K the value of resistance increases with decreasing temperature like for a semiconductor.
The current induced Coulomb oscillations are observed near 250 K in heating run. The total variation of the resistance is not large and the resistivity is of the order of 200 μΩ-cm. We observed two jumping up steps around 50 K. When temperature decreases still more, the resistance curve begins to drop abruptly from 10 K. The heating run from 6 K almost follows on the cooling run's trace except resistance variation near 250 K, i.e., the current induced Coulomb oscillation. **Figure 2** shows the resistance dropping behavior in the low temperature area. The decrease from 10 K to 3 K is about 13 % and the following abrupt variation was observed below 3 K. The resistance-zero temperature was found to be 2.1 K. This curve shows a behavior as if there are two kind of transition, as shown in the figure. We measured the temperature dependence of magnetization under 50 Oe in order to compare with the

† Received on November 1, 2010
* Graduate School, Osaka University, Osaka, Japan
** JWRI, Osaka University, Osaka, Japan

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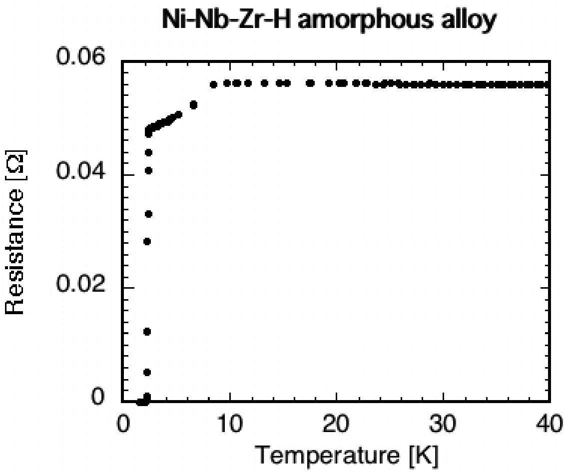

Fig. 2 Resistance variation in low temperature area.

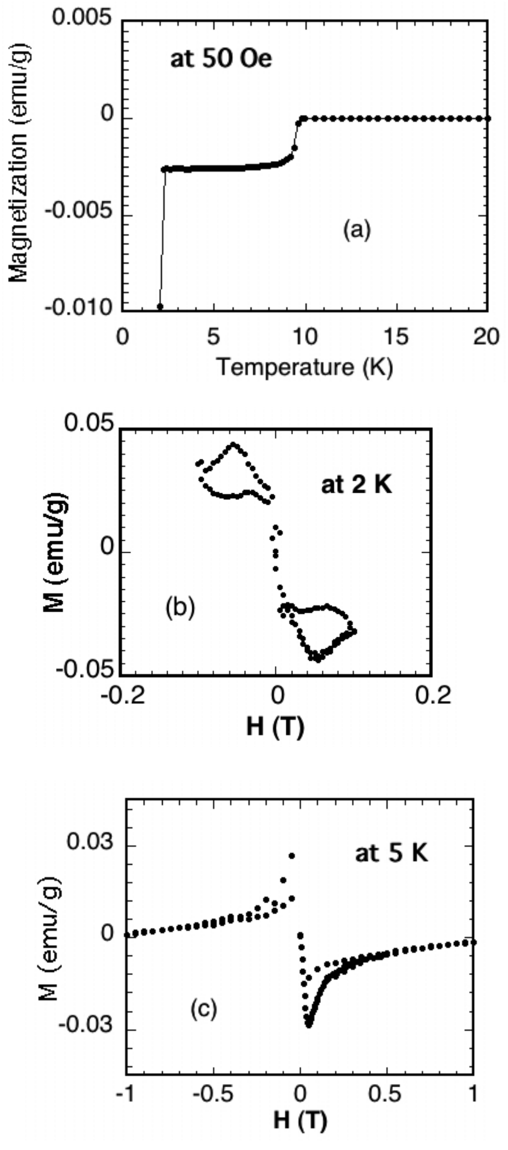

Fig. 3 (a) Magnetization vs. temperature curve under 50 Oe, (b) Magnetization loop at 2 K.

resistance measurements. A temperature variation of the magnetization at 50 Oe and the magnetization loop at 2 K and 5 K are shown in **Fig. 3**(a), (b) and (c), respectively. The magnetization at higher temperature than 10 K shows a small positive value due to Pauli paramagnetism. However, in a lesser temperature range than 10 K, the magnetization reveals a negative value and shows two steps of stages. Taking the temperature dependence of electrical resistance and magnetization into consideration, there are two peculiar regions, 2.1 K $<T<$ 10 K and T $<$ 2.1 K, for superconductivity. Magnetic loop at 2 K shows a complicated aspect in comparison with one at 5 K. In other words, the behavior of low temperature shows the aspect of the loop with plural superconducting phases. We measured the magnetoresistance effect at 4.2 K in **Fig. 4.** The measured curve shows a small abnormal peak at 4 T and then saturates, while the nonhydrogenized sample does not show the irregular peaks. A similar peak was also observed in a temperature dependent resistance curve.

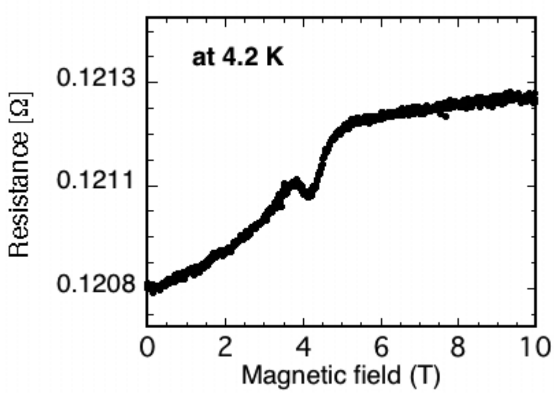

Fig. 4 Magnetoresistance effect at 4.2 K.

4. Conclusions

We measured temperature- and magnetic field dependences of superconductivity for $(Ni_{0.36}Nb_{0.24}Zr_{0.40})_{100-y}H_y$ glassy alloys in the temperature region from 300 K to 2 K and in the magnetic field from 0 T to 10 T. There are possibilities that the specimens have two kind of superconductive phase with transition temperatures of 2.1 K and 10 K. The latter phases showed an irregular peak at 4T.

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