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Giant magnetoresistance in Au-rich Co–Au and Fe–Au bulk granular alloys

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Abstract. Magnetoresistance (MR) has been investigated for Au-rich Co–Au and Fe–Au bulk aged granular alloys. A negative isotropic MR is observed at room temperature. In particular, the room-temperature value of MR of 5.5% in the $Co_{15}Au_{85}$ bulk granular alloy aged at 250 °C for 1 h is larger than that reported for Co/Au multilayers.

1. Introduction

Since the discovery of the giant magnetoresistance (GMR) in the Fe/Cr metallic superlattice [1], a considerable number of studies have been made on the GMR in the various metallic superlattices. During the last 2 years, GMR in NM-rich TR–NM (TR ≡ Fe, Co, Ni and their alloys; NM ≡ non-magnetic metals and alloys) films with nanostructured magnetic precipitates produced by sputtering have been found [2–8]. The GMR is considered to originate in spin-dependent scattering mainly at the interfaces between the magnetic layers and the non-magnetic layers, or between the magnetic granules and the matrix [7, 8]. The scattering is enhanced when the magnetic regions are polarized in different directions. The resistance of the magnetic layers with antiferromagnetically coupled moments or the various granules with randomly oriented moments is reduced by applying an external magnetic field because of the alignments of moments.

Recently, it has been reported that heterogeneous Cu-rich Co–Cu alloys produced by liquid quenching also exhibit a GMR [9–12]. In supersaturated solid solutions of TR–NM alloy systems having a miscibility gap, a GMR could be obtained in the process of phase separation even in the bulk. Au-rich Co–Au and Fe–Au alloys have a solid solution up to 23.0 at.% Co at 996.5 K and 74 at.% Fe at 1173 K, respectively, and they have a miscibility gap at low temperatures in the equilibrium state [13, 14]. Therefore, these two alloy systems are suitable for the GMR effect in the bulk state. Although a GMR of a few per cent is observed at room temperature for the Co/Au and Fe/Au multilayers [15, 16], no studies have attempted to obtain a GMR in Co–Au and Fe–Au bulk granular alloys. Bulk alloys with a high magnetoresistance (MR) ratio would open up the prospect of new applications of GMR at high currents in electrical engineering. Furthermore, such bulk alloys have some merits for ease of fabrication and also for fundamental research such as neutron scattering and atom-probe field ion microscopy.

In the present paper, we deal with the MR of Au-rich Co–Au and Fe–Au bulk alloys aged under various conditions.

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2. Experimental procedures

The alloy ingots were prepared using 99.9% pure metal elements (Co, Fe and Au) by arc melting in an argon gas atmosphere. The as-cast samples were cold rolled to reduce the thickness of the specimen with a reduction rate of 50–90%. These sample plates of size 50 mm × 10 mm × 0.1 mm were solution treated in a vacuum-sealed quartz tube at 950 °C for 120 min and quenched into water; they were then subjected to aging at 200–500 °C for 60–170 min and finally quenched again into water.

The structures of the as-quenched and aged specimens were examined by x-ray diffraction using Cu Kα radiation and transmission electron microscopy (TEM). Magnetization measurement up to 15 kOe was made at room temperature (RT) using a vibrating-sample magnetometer. The MR was measured up to 15 kOe at RT by a DC four-point probe method. The value of $\Delta\rho/\rho$ is calculated from the following equation:

$$\frac{\Delta\rho}{\rho} = \frac{\rho(H=0) - \rho(H=15\ \text{kOe})}{\rho(H=15\ \text{kOe})}. \tag{1}$$

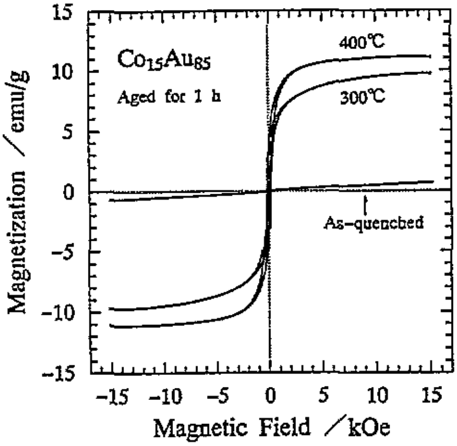

Figure 1. Magnetization curves of Co$_{15}$Au$_{85}$ bulk alloys as quenched and aged at different temperatures for 1 h.

3. Results and discussion

The magnetization curves for the Co$_{15}$Au$_{85}$ alloys as quenched and aged at different temperatures for 1 h are shown in figure 1. In the as-quenched state, the magnetization curve shows a paramagnetic behaviour at RT, which means that Co atoms completely dissolved in the Au matrix. On the other hand, the magnetization curves of the samples aged at and above 300 °C are saturated more easily, which means that a Co-rich phase with a higher Curie temperature than RT precipitates in the Au matrix. These results are reasonable because each solubility limit for Co–Au alloys even at 500 °C in the equilibrium state is 0.2 at.% Au and 0.3 at.% Co [13].

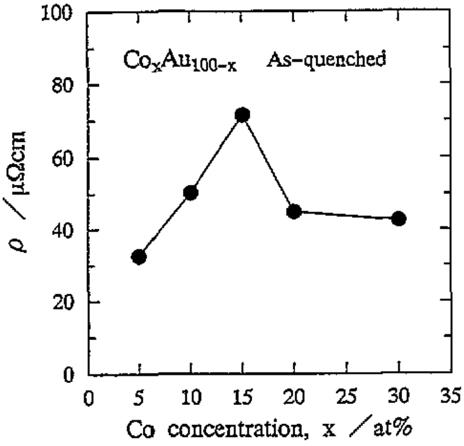

Figure 2. Concentration dependence of resistivity ρ for as-quenched Co_xAu_{100-x} bulk alloys.

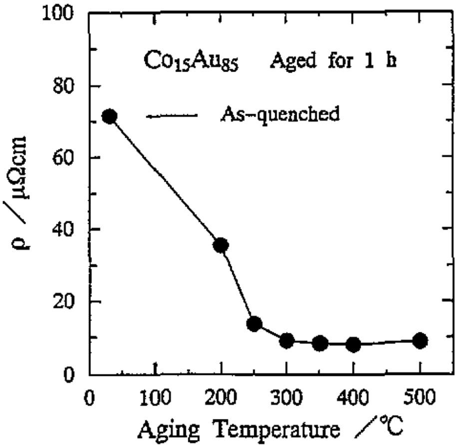

Figure 3. Aging temperature dependence of the resistivity ρ for $Co_{15}Au_{85}$ bulk alloys as quenched and aged for 1 h.

Figure 2 shows the concentration dependence of the resistivity for the as-quenched Co_xAu_{100-x} alloys. The increase in ρ up to $x = 15$ in the as-quenched state suggests that the samples have a solid solution, being in accord with the magnetization curve in figure 1.

Figure 3 shows the aging temperature dependence of resistivity ρ for the $Co_{15}Au_{85}$ alloys as quenched and aged for 1 h. With increasing aging temperature, in addition to the decrease in defects introduced by quenching, the decomposition to the Au-rich matrix with a low value of resistivity and magnetic granules causes a decrease in ρ.

Negative isotropic MRs were observed in the $Co_{15}Au_{85}$ alloys aged at 300 °C for 1 h, as shown in figure 4. It should be noted that the MR ratio of 5% is larger than that for the Co/Au multilayers [15] and that these negative MRs for both $\boldsymbol{H} \parallel \boldsymbol{i}$ and $\boldsymbol{H} \perp \boldsymbol{i}$ (where $\boldsymbol{H}$ means the magnetic field direction and $\boldsymbol{i}$ the current direction in plane) distinguish GMR from the ordinary and anisotropic MRs, regardless of their magnitude. Since the MR for Fe/Cr multilayers is so large [1], it has been widely referred to as the GMR. However,

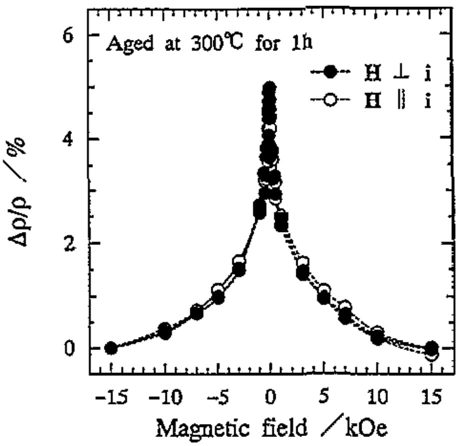

Figure 4. Field dependence of the resistivity for $Co_{15}Au_{85}$ bulk alloy aged at 300 °C for 1 h.

what distinguishes the GMR form the ordinary anisotropic MRs is not its magnitude but the fact that it is negative in both the longitudinal and the transverse field directions in the heterogeneous structure [5]. Although negative isotropic MRs have been observed for Kondo alloys [16], spin glasses and so on [17–19] by suppression of spin fluctuation and/or spin-disorder scattering, the GMR effect is related to the change in the moment arrangement in the magnetic layers and granules with the magnetic field.

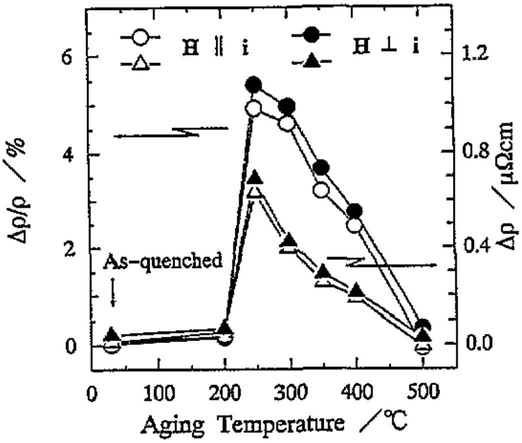

Figure 5. Aging temperature of the change $\Delta\rho = \rho(H = 0\ \text{kOe}) - \rho(H = 15\ \text{kOe})$ in resistivity and the MR ratio $\Delta\rho/\rho$ for $Co_{15}Au_{85}$ bulk alloys as quenched and aged for 1 h.

Figure 5 shows the aging time dependence of the change $\Delta\rho = \rho(H = 0\ \text{kOe}) - \rho(H = 15\ \text{kOe})$ in the resistivity and the MR ratio $\Delta\rho/\rho$ for the $Co_{15}Au_{85}$ alloys as quenched and aged for 1 h. The values of $\Delta\rho/\rho$ shows a maximum on aging. The largest value of 5.5% is obtained on aging at 250 °C for 1 h owing to both the decrease in ρ and the increase in $\Delta\rho$.

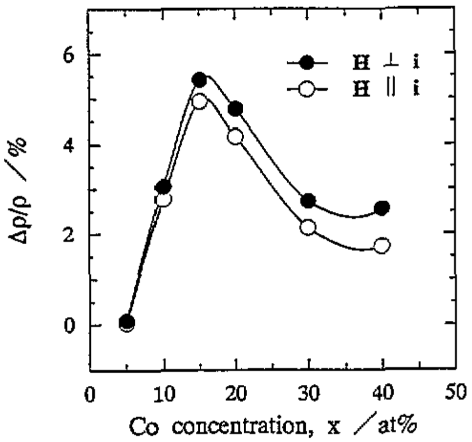

Figure 6. Concentration dependence of the MR ratio for Co_xAu_{100-x} bulk alloys aged at optimum conditions around 300 °C for 1 h.

As shown in figure 6, the concentration dependence of the MR ratio of the Co_xAu_{100-x} alloys at the optimum aging around 300 °C for 1 h shows a maximum at 15 at.% Co. This concentration is consistent with the model proposed by Zhang [7], i.e. the MR ratio increases with increasing volume fraction of the magnetic granules. The ratio decreases with increasing granule size if we assume that large magnetic particles are formed beyond the percolation limit, which result in a decrease in the interface area between magnetic granules and matrix, and a decrease in the portion of random orientation of magnetic moment due to ferromagnetic coupling. The change from the superparamagnetic state to ferromagnetic coupling among magnetic granules with increasing x is consistent with the increase in the difference between the MRs for $H \perp i$ and $H \parallel i$, which suggests an increase in the anisotropic MR. The change is also confirmed in the concentration dependence of the magnetization curves for the Co_xAu_{100-x} bulk granular alloys. An improvement in the MR ratio should still be possible by selecting appropriate conditions such as the composition and heat treatments.

Similar behaviour of the MR is observed in bulk Au-rich Fe–Au alloys. For example, bulk $Fe_{40}Au_{60}$ alloy aged at 500 °C for 1 h shows a GMR of 2% at RT. The value is the same magnitude as that for Fe/Au multilayers [20].

In conclusion, GMR has been observed even for bulk samples such as Au-rich Co–Au and Fe–Au aged granular alloys. In particular, it should be emphasized that the RT MR value of 5.5% for the $Co_{15}Au_{85}$ granular aged alloy is larger than that reported for Co/Au multilayers.

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