Tunnel magnetoresistance of disordered, low-moment Co_2MnSi Heusler alloy thin films

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Thin films of $Co_{(1-x-y)}Mn_xSi_y$ ($x=0.18-0.26$ and $y=0.23-0.29$) were deposited from elemental targets onto thermal-oxidized Si substrates at room temperature using dc magnetron cosputtering. The as-deposited films appear to be amorphous showing no distinguishable peaks in x-ray diffraction, and the magnetic moment at room temperature is nearly zero. Despite of its low magnetic moment (less than $0.001\mu_B$ per formula), the disordered Heusler magnetic tunnel junctions show tunnel magnetoresistance ratios as large as 10% at room temperature. The dependence of the tunnel magnetoresistance ratio on compositions x and y is also reported. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1846431]

I. INTRODUCTION

One of the properties of materials of interest in spin electronics is half-metallicity. In a half-metal, conduction electrons are 100% spin polarized at $T=0$ due to a gap at the Fermi level in the minority- (or majority-) spin band and a finite density of states in the majority (or minority) band. The first material, which was identified as a half-metal, was the half-Heusler alloy NiMnSb studied by de Groot *et al.*[1] in 1983. Although half-Heusler alloys like NiMnSb have attracted a lot of interest, the other family of Heusler compounds, the full-Heusler alloys, has been studied much more extensively of late. The full-Heusler alloys crystallize in the $L2_1$ structure that comprises four fcc sublattices. Ziebeck and Webster[2] synthesized full-Heusler alloys containing Co, and Ishida *et al.*[3] predicted the half-metallicity of Co_2MnSi.

Following the Jullière model, a high tunnel magnetoresistance (TMR) could be expected when half-metals are incorporated as electrodes in magnetic tunnel junctions (MTJs). Ideally, the 100% spin polarization of the half-metals can lead to a TMR ratio of at least 100%, which would be very suitable for device applications. However, up to now, attempts at building MTJs with half-metals have reported lower TMR values.[4–6] A successful attempt was made by Kämmerer *et al.*, who reported a 61% TMR at 10 K for Co_2MnSi.[4] The Other was by Inomata *et al.*[5] for $Co_2CrFeAl$. They attributed the relatively low TMR value of 27% at 5 K to the B_2 structure of the film and consequent low magnetic moment. According to electronic structure calculations, the half-metallicity of the full-Heusler alloys X_2YZ is only anticipated in the ordered phase $L2_1$.[7] Miura *et al.*[8] recently calculated the electronic and magnetic structures of disordered $Co_2Cr_{0.6}Fe_{0.4}Al$, in which they reported that the XY disorder completely destroys high spin polarization. In the case of the YZ disorder, the spin polarization is less sensitive to the level of disorder, but falls to 0.77 for the B_2 structure. A similar theoretical investigation was also carried out by Picozzi *et al.*[9] for Co_2MnGe and Co_2MnSi, showing that the X antisites destroy the half-metallicity and reduce the spin polarization drastically.

Here we present the structural and magnetic properties of $Co_{(1-x-y)}Mn_xSi_y$ ($x=0.18-0.26$ and $y=0.23-0.29$) films. All the as-deposited films show no distinct Bragg reflections in x-ray diffraction and their magnetization is over ten times smaller than that of the annealed samples. It is shown that magnetic tunnel junctions with a disordered Co_2MnSi bottom electrode show a magnetoresistance as large as 10% at room temperature.

II. EXPERIMENT

Films were grown by dc magnetron cosputtering in a Shamrock tool from three elemental targets on glass substrates or thermal-oxidized Si substrates. The sputtering tool consists of three load lock vacuum chambers and the process chamber is always kept in a pressure below 1×10^{-7} Torr. All depositions were carried out at room temperature. The sputtering system has six targets arranged in a planetary configuration, and the substrates rotate along with the planetary movement at 44 rpm. Sputtering rates of less than 0.03 nm/s from each target allowed the uniform deposition of composite films. The film compositions were controlled by tuning the sputtering rate of each element individually, and were checked by energy dispersive x-ray analysis in a scanning electron microscope using films deposited on MgO. Structural and magnetic characterizations were performed by x-ray diffraction and in a superconducting quantum interference device (SQUID) magnetometer, respectively. Some of the samples were annealed at 500 °C for 2 h in a separate vacuum chamber after deposition.

Tunnel functions were fabricated in a crossgeometry using metal shadow masks with widths of 100–300 μm. The stack is $Si/SiO_2/Co_2MnSi$ 50 nm/Al 1.4 nm oxidized/Co 3 nm/IrMn 20 nm/Ta 5 nm. Al thin films were oxidized by a 90-s exposure to oxygen plasma from an inductive coupled plasma coil at a 1 mTorr O_2 partial pressure and a 20 W rf power.

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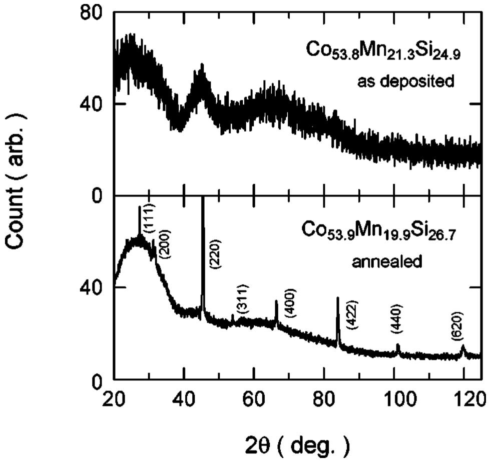

FIG. 1. X-ray diffraction patterns for $Co_{(1-x-y)}Mn_xSi_y$: thin films. (a) as-deposited and (b) annealed.

III. STRUCTURE AND MAGNETIZATION

In Fig. 1, we compare the x-ray diffraction patterns of as-deposited and annealed $Co_{(1-x-y)}Mn_xSi_y$ films. The film thickness is 340 nm. Regardless of the composition, all the as-deposited samples show no distinct peaks, exhibiting an essentially amorphous phase. After annealing, the films are polycrystalline with a strong (110) texture. Only the samples in the narrow range of Si compositions ($y=0.255-0.278$) show both (111) and (200) peaks that correspond to the $L2_1$ ordered phase. The magnetization curves of a $Co_{52.6}Mn_{20.2}Si_{27.2}$ film are shown in Fig. 2. After annealing, the sample exhibits the $L2_1$ ordered phase in x-ray diffraction, and shows an almost square in-plane hysteresis loop at 5 K with a coercivity less than 5 mT. Even though the film shows soft magnetic behavior, which is characteristic of the ordered Heusler alloy, the remanent magnetization of 535 kA/m ($2.5\mu_B$ per formula) is still 50% lower than the bulk value of 1040 kA/m ($5.1\mu_B$). The lower magnetization value can be explained by taking into account the off stoichiometry of the film. By contrast, as-deposited, the film has a remanent magnetization of only 76.5 kA/m ($0.33\mu_B$) at 5 K, which falls off to only 0.18 kA/m at room temperature.

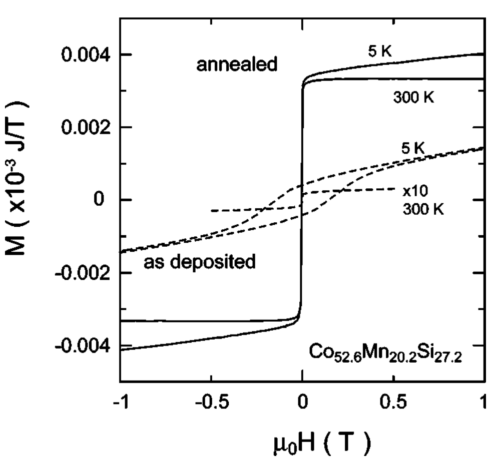

FIG. 2. Magnetization curves for a $Co_{52.6}Mn_{20.2}Si_{27.2}$ thin film. The broken and solid lines represent the magnetization curve taken at as-deposited and after annealing, respectively.

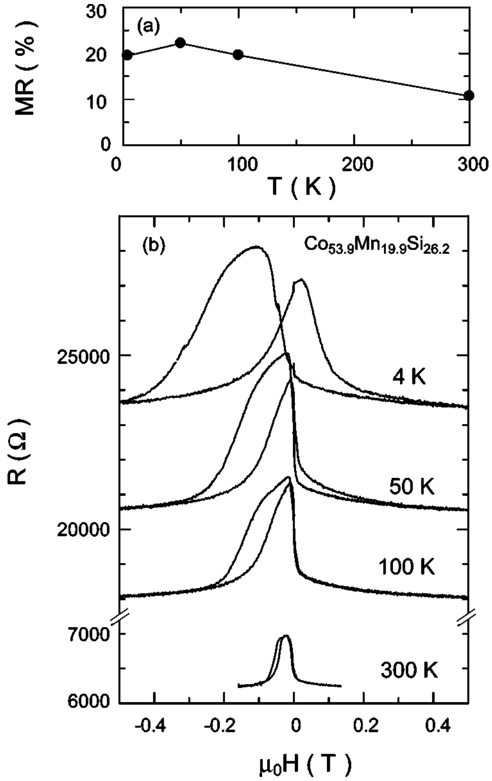

FIG. 3. (a) Temperature dependence of the tunnel magnetoresistance of a disordered Heusler tunnel junction. (b) Magnetoresistance curves of the junction.

IV. TUNNEL MAGNETORESISTANCE OF DISORDERED FILMS

It is surprising that even though the disordered films have very low magnetic moments, tunnel junctions made with these disordered films show magnetoresistance (TMR) ratios as high as 10% at room temperature. Figure 3(a) shows the temperature dependence of the TMR ratio for a junction with a $Co_{53.9}Mn_{19.9}Si_{26.2}$ film as a bottom electrode. At room temperature, the device shows 10.7% TMR with separate switching of the free and pinned layers. The value increases at lower temperature, reaching a maximum value of 22.7% at 50 K. The decline of TMR from 50 to 4 K is explained by the deterioration of the separate switching due to the large coercivity of the Heusler film itself, as shown in Fig. 3(b). The coercivity at 4 K agrees well with SQUID measurements. The increase of TMR at low temperature does not follow the increase of magnetic moment. This implies that magnetic moment does not directly determine the spin polarization of the conduction electrons in this case. In Fig. 4, TMR ratios as a function of film composition are summarized. The values displayed on the figure are each an average based on 8 to 36 devices. The stoichiometric composition is shown as a filled black circle. The data clearly show that the TMR has a sharp composition dependence. The TMR quickly falls off when the Co content exceeds 0.55, which is contrary to our initial expectation. The TMR appears less sensitive to the Mn content. If the amorphous phase of Co_2MnSi could be simply regarded as a dilution of Co by Si and Mn, the TMR might be expected to increase as the Co content increases, or to be rather insensitive to cobalt con-

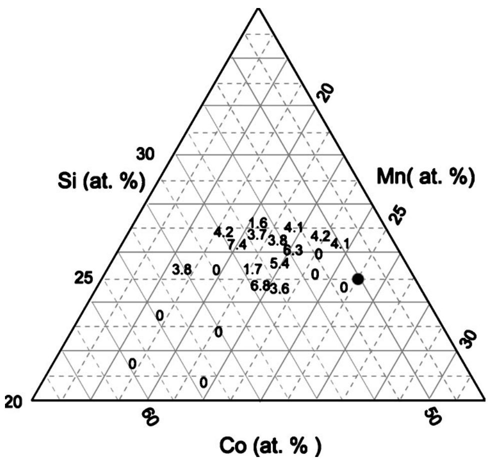

FIG. 4. Ternary diagram of tunnel magnetoresistance for devices made with disordered Heusler alloys. The solid circle indicates the stoichiometric composition.

tent. However, experimental results clearly indicate the composition sensitivity of the TMR of the disordered Co_2MnSi. The small moment can be understood in terms of the antiparallel coupling of the Co and Mn amorphous subnetworks. The Co/Mn majority density of states may have a spin gap, which disappears beyond the threshold concentration.

V. CONCLUSION

Thin films of the $Co_{(1-x-y)}Mn_xSi_y$ family ($x=0.18-0.26$ and $y=0.23-0.29$) exhibit substantial spin polarization in the amorphous state, in a range of composition where they have very little net magnetization. The small magnetic moment at room temperature, less than $0.001\mu_B$, is explained in terms of the antiparallel coupling of the Co and Mn amorphous subnetworks. The Co subnetwork is strongly ferromagnetic, and the atoms of the Mn subnetwork couple antiparallel to their cobalt neighbors. Our results show that it is possible to have spin polarization in an amorphous film with almost no net magnetization, which may be useful for some applications.

ACKNOWLEDGMENT

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