Q-17

Spontaneous Hall Effect in Amorphous Tb-Fe and Sm-Fe Thin Films

T. W. Kim[1], S. H. Lim[2] and R. J. Gambino[3]

[1] *Microelectronics Lab., Samsung Advanced Institute of Technology,*
P. O. Box 111, Suwon, Kyonggi-do, 440-600, Korea
[2] *Thin Film Technology Research Center, Korea Institute of Science and Technology,*
P. O. Box 131, Cheongryang, Seoul, 130-650, Korea
[3] *Dept. of Materials Science and Engineering, SUNY at Stony Brook,*
New York, 11794, USA

Abstract

The spontaneous Hall effect in amorphous Tb-Fe and Sm-Fe thin films, which possess excellent magnetic softness, is investigated in this work to seek a possibility of practical applications of these thin films as sensors. The resistivity of Tb-Fe thin films ranges from 180 to 250 μΩcm as the Tb content varies from 35 to 46 at.%. Tb-Fe thin films show negative Hall resistivity ranging from –7.3 to –5.0 μΩcm in the same composition range, giving the normalized resistivity ratio in the range of –4.1 to –2.0 %. On the other hand, the resistivity of Sm-Fe thin films ranges from 150 to 166 μΩcm as the Sm content varies from 22 to 31 at.%. Sm-Fe thin films show positive Hall resistivity which varies from 7.1 to 2.8 μΩcm in the same composition range, giving the normalized resistivity ratio in the range of 4.8 to 1.7 %. These values are significantly high compared with the values of other R-T alloys, Tb-Co alloys for example, where the highest reported value is 2.5 %. Between the two different sets of samples, Tb-Fe thin films with perpendicular anisotropy are considered to be more suitable for practical applications, since saturation is reached at a low magnetic field, approximately 2 kOe in a $Tb_{35.1}Fe_{64.9}$ thin film, for example.

Keywords: spontaneous Hall effect, resistivity, amorphous Sm-Fe alloys, amorphous Tb-Fe alloys, sputtered thin films

S. H. Lim (Corresponding Author)
Thin Film Technology Research Center, Korea Institute of Science and Technology, P. O. Box 131, Cheongryang, Seoul, 130-650, Korea.
Tel +82-2-958-5415, Fax +82-2-958-6851, Email sangho@kist.re.kr

1. Introduction

Many unusual electronic transport properties are observed in amorphous R-TM alloys (where R and TM denote, respectively, rare earths and 3d transition metals), and one notable example is a large spontaneous Hall effect [1]. The investigation of the Hall effect in R-TM alloys is of great theoretical importance, since it provides us a means of understanding the electronic transport in these alloys. Furthermore, due to their large spontaneous Hall effect [1], amorphous R-TM alloys are also of significant practical importance. In practical applications, it is very important for the materials to have a large Hall effect at a low magnetic field. Recently, magnetostrictive thin films of amorphous Tb-Fe and Sm-Fe alloys with excellent soft magnetic properties were developed by one of the authors, and they were found to exhibit a very large strain at a low field [2,3]. The aim of the present study is to examine the spontaneous Hall effect of amorphous Tb-Fe and Sm-Fe thin films and to seek a possibility of practical applications of these soft magnetic R-TM thin films. The thin films of the alloys investigated are Tb_xFe_{100-x} with x= 35.1, 37.5, 41.3 and 45.5, and Sm_yFe_{100-y} with y=21.6, 23.8, 28.7 and 30.8 (in at.%).

2. Experimental Details

Thin films with a thickness of about 1 μm were coated by rf magnetron sputtering on Si (100) substrates. The shape of the thin films was circular with a diameter of 10 mm. A composite target consisting of a pure Fe disc (4 inches in diameter) and R chips was used. Argon was used as the sputtering gas. The sputtering pressure was in the range of 1 to 10 mTorr. Other sputtering conditions used in this work were: a base pressure of below 7×10^{-7} Torr, a target to substrate distance of 60 mm and an rf input power of 300 W. The film thickness was measured by using a stylus-type surface profiler. The film composition was determined by electron probe microanalysis. The microstructure was observed by x-ray diffraction with Cu Kα radiation. The microstructure of all the alloy thin films investigated in this work was observed to consist of a single amorphous phase. Magnetic properties were measured by using a vibrating sample magnetometer with a maximum magnetic field of 15 kOe. The transport properties including the Hall effect were measured by using the van der Pauw method at magnetic fields up to 6 kOe [4]. The field was applied in the direction perpendicular to the film plane. Four silver paste contacts were formed at right angles. For resistivity measurements, the current was applied through two adjacent contacts and the voltage was measured at the rest two adjacent ones. In the case of Hall effect measurements, however, oppositely positioned contacts were used for current and voltage. All the measurements were done at room temperature.

3. Results and Discussion

The total Hall resistivity (ρ_H) of spontaneously magnetized materials consists of two components: namely, the ordinary (ρ_o) and the spontaneous (or extraordinary) (ρ_s) components [1]. In the CGS unit convention, the following relation exists;

$$\rho_H = R_o H + R_s 4\pi M_s$$

where R_o and R_s are, respectively, the ordinary Hall coefficient and the spontaneous Hall coefficient, and M_s is the saturation magnetization. The ordinary Hall effect arises from the Lorentz force acting on the moving electrons, and the spontaneous Hall effect results from asymmetric (skew or side jump) scattering of the conduction electrons by the magnetic atoms. Since, in a magnetic material, the magnitude of R_o is known to be much smaller than that of R_s, the Hall resistivity of a magnetic material is essentially given by the last term of the equation ($R_s 4\pi M_s$).

In Fig.1 are shown the ρ_H –H loops for the thin films of $Tb_{35.1}Fe_{64.9}$ and $Sm_{21.6}Fe_{78.4}$ alloys. No correction was made with respect to the demagnetization field. In the case of the Tb-Fe thin film, saturation is reached at about 2.5 kOe, and an appreciable amount of hysteresis is observed, being particularly true near saturation. The ease of saturation and the existence of hysteresis can be understood from the fact that the Tb-Fe thin film possesses the perpendicular anisotropy [5]. Another important point to note is that the way ρ_H varies with H is opposite to that observed in an ordinary M-H loop. Specifically, the value of ρ_H decreases (increases) with increasing (decreasing) H, indicating that the sign of R_s is negative. This negative R_s can be explained as follows. In a model proposed by McGuire et al.[4], both R and TM sublattices contribute to the total Hall resistivity. Tb and Fe elements possess negative and positive Hall voltages, respectively. It is noted here that an extensive study on amorphous R-M alloys (where M is a non-magnetic element) indicates a positive Hall voltage for light rare earths (such as Sm), and a negative Hall voltage for heavy rare earths (such as Tb) [6]. Amorphous Tb-Fe alloys exhibit the ferrimagnetic spin structure [1]. The Tb content of the $Tb_{35.1}Fe_{64.9}$ alloy is higher than that of the compensation composition. The magnetization of the composition is dominated by the Tb magnetization (the absolute value of Tb magnetization is greater than that of Fe). At the composition, the negative value of Tb contributes a minus Hall voltage and the positive value of Fe also contributes a minus Hall voltage because of the ferrimagnetic spin structure. It is therefore expected that both elements generate a negative Hall voltage. The Hall resistivity at saturation (namely, ρ_H) is –7.3 μΩcm. A slight increase of the Hall resistivity is observed even after saturation, and this increase is considered to be mainly due to the contribution from the ordinary Hall effect. It is quite obvious from the ρ_H –H loop that the ordinary Hall effect is much smaller than the spontaneous effect. The shape of the ρ_H –H loop for the Sm-Fe thin film is very different from that for the Tb-Fe thin film. In this case, saturation is not reached with the maximum applied field of 6 kOe and no hysteresis is observed. Furthermore, the sign of R_s is positive. The first two behaviors are due to the fact that the Sm-Fe thin film exhibits in-plane anisotropy. The absence of hysteresis indicates that the magnetization mainly occurs by reversible spin rotation. This magnetization behavior would be expected when most of the spins reside in the direction perpendicular to the applied field, and this is in agreement with the previous observation that amorphous Sm-Fe thin films possess a very well developed in-plane anisotropy [7]. The positive R_s value can be explained from the two facts that (1) the spin directions of Sm and Fe in ferromagnetic Sm-Fe alloys are parallel to each other [1] and (2) both elements possess positive Hall voltages [6]. The linear increase of ρ_H is expected until the applied field is approximately equal to $4\pi M_s$ (7.92 kOe in the $Sm_{21.6}Fe_{78.4}$ thin film), which is the field necessary to overcome the demagnetization field in the perpendicular direction. From this linear approximation, the magnitude of ρ_H is estimated to be 7.12 μΩcm.

In Fig.2 are shown the results for ρ_H as a function of composition for both Sm-Fe and Tb-Fe alloy thin films. Since saturation of the Sm-Fe thin films is not reached with the maximum magnetic field applied in this work (6 kOe), the results for these alloys are estimated from the linear approximation, as was discussed earlier. In both thin films, the *absolute* magnitude of ρ_H decreases with increasing R content, the compositional dependence of ρ_H being much greater in Sm-Fe thin films. In the composition range investigated in this work, the value of ρ_H varies from –7.3 to –5.0 μΩcm in the case of Tb-Fe thin films, and it is in the range of 7.12 to 2.8 μΩcm in the Sm-Fe thin films.

The Hall effect can better be characterized by the ratio ρ_H/ρ (or also frequently called the tangent of the Hall angle) (where ρ is the electrical resistivity), since both ρ_H and ρ arise from the same atomic scattering sources [4]. The composition dependence of the Hall angle (ρ_H/ρ) provides evidence of the effects of the exchange interaction on the

Hall scattering. In R metals and alloys the RKKY exchange mechanism is believed to be dominant. This exchange is between conduction electron spins and the localized spins of the R. The conduction electrons are polarized by this exchange and the strength of the exchange is related to the Curie temperature or Curie Weiss θ [8]. The increase in spin polarization of the conduction electron enhances the intrinsic spin-orbit coupling [8]. In Fig. 3 are shown the results for the compositional dependence of ρ. In both Tb-Fe and Sm-Fe thin films, the magnitude of ρ increases with increasing R content, the increase being steeper and the value itself being larger in the Tb-Fe thin films than in the Sm-Fe ones. In the present composition range, the electrical resistivity of the Tb-Fe thin films varies from 180 to 250 μΩcm, while that of the Sm-Fe thin films ranges from 150 to 166 μΩcm. The R content dependence of ρ can be explained by an increased mixing and hence increased electronic scattering by atoms with the increase of the Tb content in the composition range below 50 at.%. It is well-known that, in amorphous and liquid alloys, the value of ρ reaches a maximum near 50:50 composition where the electron scattering is highest [1]. The higher ρ values of the Tb-Fe thin films can thus be explained in terms of the atomic configuration (mixing). However, an intrinsic chemical effect cannot be ruled out, since the value of ρ in the Tb-Fe thin films is much higher than that extrapolated from the Sm-Fe thin films with low R contents.

It is now possible to obtain the results for the ratio ρ_H/ρ from those for ρ_H (Fig.2) and ρ (Fig.3), and these results are shown in Fig.4. It is seen from the figure that the compositional dependence of the ratio ρ_H/ρ is steeper than that of ρ_H, since the denominator (ρ) increases with increasing R. This is particularly true for the Tb-Fe thin films due to a large compositional dependence of ρ. In the present composition range, the ratio of the Tb-Fe thin films varies from –4.06 to –2.0 %, while that of the Sm-Fe thin films is in the range of 4.75 to 1.69 %. The understanding of the R content dependence of ρ_H/ρ (also ρ_H) can only be possible when the information on the contribution to the total (spontaneous) Hall effect from the two sublattices is known. Since the magnetic structure and the sign of the spontaneous Hall effect of each sublattice in amorphous R-TM alloys indicate that the total spontaneous Hall effect is simply the sum of the R and TM sublattice contribution, the equation for ρ_H can be re-written as;

$$\rho_H = R_o H + R_s^R 4\pi M_R + R_s^{TM} 4\pi M_{TM}$$

The notations in the equation are obvious. The decrease of the absolute magnitude of ρ_H/ρ (also ρ_H) with increasing R content obviously indicates that the contribution to the spontaneous Hall effect from the R sublattice is much higher than that of TM one, namely,

$$\left|R_s^R 4\pi M_R\right| < \left|R_s^{TM} 4\pi M_{TM}\right|.$$

It is important to point out here that, due to the nature of the electronic shell structure, the TM sublattice term is a sensitive function of the composition, while the R sublattice term remains constant. Specifically, a charge transfer to the unfilled 3d shell of Fe from the rare earth occurs by alloying causing the reduction of the sublattice magnetization of Fe and hence the spontaneous Hall effect [4] with the increase of the R content in the alloy. This charge transfer related effect plays a role of enhancing the compositional dependence of ρ_H/ρ (also ρ_H).

The present values of ρ_H/ρ are considered to be significantly high compared with other R-TM alloys, amorphous Tb-Co alloy thin films, for example, where the highest value is 2.5 % at the Tb content of 19 at.% [8]. Furthermore, the present thin films exhibit excellent magnetic softness. This can clearly be demonstrated by a comparison of the coercivity values between the thin films of Tb-Fe and Tb-Co, both of which possessing the perpendicular anisotropy; the coercivity of the Tb-Fe thin film (approximately 50 Oe, see Fig.1) is significantly lower than that of the Tb-Co one (2000 Oe) [8]. Between the two different thin film systems, the Tb-Fe system is considered to be more suitable for practical applications than the Sm-Fe one due to its much lower saturation magnetic field, combined with a high Hall effect.

4. Conclusions

Magnetostrictive thin films of amorphous Tb-Fe and Sm-Fe alloys with excellent soft magnetic properties were recently developed by one of the authors, and they were found to exhibit a very large strain at a low field. The aim of the present study was to examine the spontaneous Hall effect and to seek a possibility of practical applications of these soft magnetic R-T thin films. In practical applications, it is important for the materials to have a large Hall voltage at a low magnetic field. Amorphous thin films were fabricated by rf magnetron sputtering. The Hall effect was measured at room temperature by using the Van der Pauw method with the magnetic field applied in the direction perpendicular to the film plane. The resistivity of Tb-Fe thin films ranges from 180 to 250 μΩcm as the Tb content varies from 35 to 46 at.%. Tb-Fe thin films show negative Hall resistivity ranging from –7.3 to –5.0 μΩcm in the same composition range, giving the normalized resistivity ratio in the range of –4.1 to –2.0 %. On the other hand, the resistivity of Sm-Fe thin films ranges from 150 to 166 μΩcm as the Sm content varies from 22 to 31 at.%. Sm-Fe thin films show positive Hall resistivity which varies from 7.1 to 2.8 μΩcm in the same composition range, giving the normalized resistivity ratio in the range of 4.8 to 1.7 %. Tb-Fe thin films with perpendicular anisotropy are considered to be more suitable for practical applications than Sm-Fe thin films possessing well-developed in-plane anisotropy. This is because saturation is reached at a low magnetic field in the case of Tb-Fe thin films

References

1. P. Hansen, in Handbook of Magnetic Materials, vol. 6, edited by K. H. J. Buschow (Elsevier Science Publishers B. V., Amsterdam, The Netherland), chap 4, 1991
2. S. H. Lim, Y. S. Choi, S. H. Han, H. J. Kim, T. Shima and H. Fujimori, IEEE Trans. Magn., vol. 33, 3940 (1997)
3. S. H. Lim, Y. S. Choi, S. H. Han, H. J. Kim, T. Shima and H. Fujimori, J. Magn. Magn. Mater., vol. 189, 1 (1998)
4. T. R. McGuire, R. J. Gambino and R. C. Taylor, J. Appl. Phys., vol. 48, 2965 (1977)
5. Y. S. Choi, S. R. Lee, S. H. Han, H. J. Kim and S. H. Lim, J. Alloys Comp., vol. 258, 155 (1997)
6. T. R. McGuire and R. J. Gambino, J. Appl. Phys., vol. 50, 7653 (1979)
7. Y. S. Choi, S. R. Lee, S. H. Han, H. J. Kim and S. H. Lim, J. Appl. Phys., vol. 83, 7270 (1998)
8. T. R. McGuire, R. J. Gambino and R. C. O'Handley, "Hall effect in amorphous metals" in The Hall Effect and Its Applications, edited by C. L. Chien and C. R. Westgate, Plenum Publishing Corp., New York, p137 (1980)
9. T. W. Kim and R. J. Gambino, J. Appl. Phys. (to be Published)

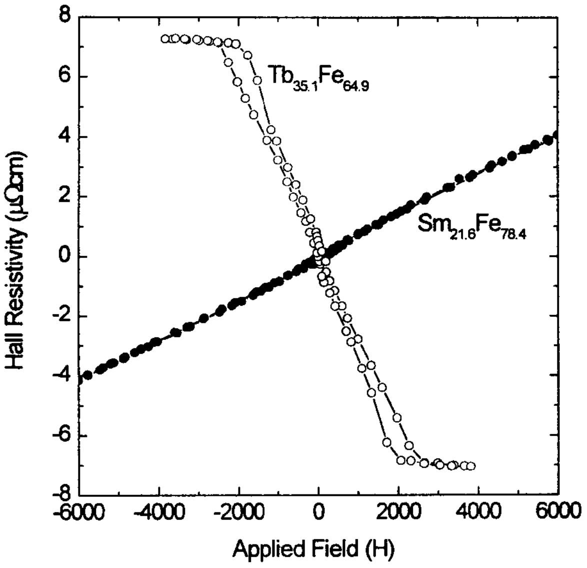

Figure 1. The ρ_H –H loops for the thin films of $Tb_{35.1}Fe_{64.9}$ and $Sm_{21.6}Fe_{78.4}$ alloys. No correction was made with respect to the demagnetization field.

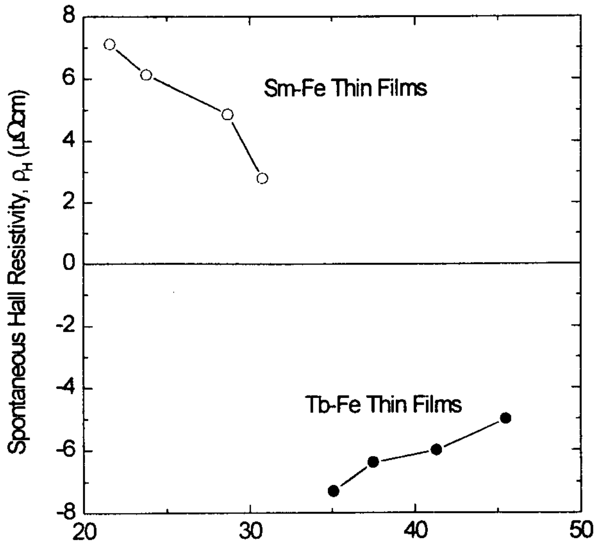

Figure 2. The value of the spontaneous resistivity (ρ_H) as function of composition for both Sm-Fe and Tb-Fe alloy thin films.

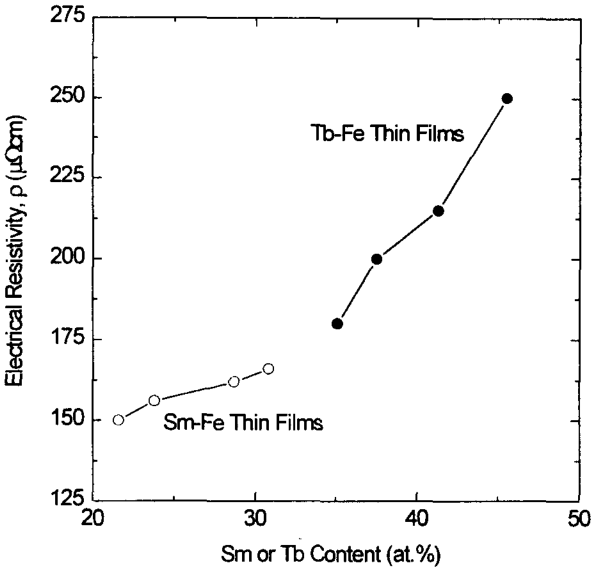

Figure 3. The value of the electrical resistivity (ρ) as a function of composition for both Sm-Fe and Tb-Fe alloy thin films.

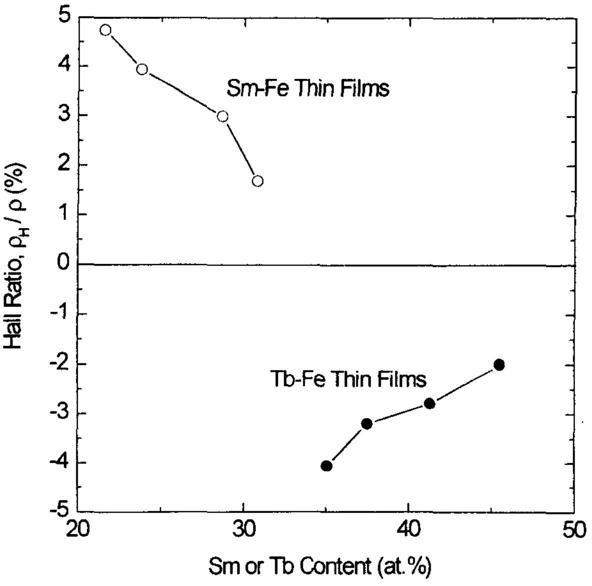

Figure 4. The value of the Hall ratio (ρ_H/ρ) as a function of composition for both Sm-Fe and Tb-Fe alloy thin films.