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Magnetic properties and switching volumes of nanocrystalline SmFeSiC films

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Systematic studies of the effects of Si addition on the magnetic and magnetization reversal properties of SmFeSiC films are presented. The magnetic switching volume and other magnetic parameters (e.g., coercivity) are strongly dependent upon the Si content. Correlations between switching volume, coercivity, and the intergrain interactions are discussed. © *1997 American Institute of Physics.* [S0021-8979(97)54408-2]

I. INTRODUCTION

Studies of nanocrystalline semihard and hard magnetic films are important both for fundamental understanding and possible applications, such as magnetic recording media, bias magnets, and exchange-spring coupled materials. It is important to have high coercivity in these films, which may be achieved by multilayer sputtering the constituent layers followed by a solid-state reaction. In recent years much attention has been focused on the preparation and study of $Sm_2Fe_{17}C_y$ bulk materials[1] and rather little work on SmFeC thin films has been reported. In our previous article,[2] we have studied the effects of preparation conditions on the magnetic properties of SmFeSiC films and optimum preparation conditions have been determined to increase the film coercivity to 7.7 kOe. However, the magnetic and magnetization reversal properties have not been investigated intensively. In the present article we will focus on analyzing the magnetic properties, especially emphasizing the effects of Si addition on the magnetic properties and switching volumes of these films.

II. EXPERIMENTS

SmFeSiC thin films were prepared by sputtering SmFe/C(Si) multilayers with a Ta underlayer onto Si substrates and subsequently annealing at 700 °C for about 5 min. The details of the preparation condition of the films can be found in Ref. 2. Compositions of films were examined with an EDX spectrometer, and the magnetic properties including the magnetization M, coercivity H_c, intergrain interaction behavior ΔM,[3] and the magnetic switching volume or magnetic grain,[4,5] were measured with an alternating gradient force magnetometer.

III. RESULTS AND DISCUSSION

It has been reported that the high coercivity in SmFeC alloys is difficult to obtain and the addition of a fourth element, e.g., Si, Ga, or Al, is helpful in overcoming this barrier.[6,7] Systematic studies of the Si addition effects on the hysteresis loops of $(Sm_2Fe_{12})_{1-x}Si_xC_y$ films are shown in Fig. 1, which gives general features of the magnetic properties. It is seen clearly that the film without Si addition displays a very narrow loop, and the loops become broad rapidly as the Si content increases from 0 to 0.072 and then remains nearly unchanged as x increases further to 0.15. Our previous article[2] has pointed out that the preparation conditions of the Ar pressure and annealing temperature also have significant effects on the magnetic properties of films. However, it should be emphasized that the appropriate Si addition is the key point to fabricate films with high coercivity; a high

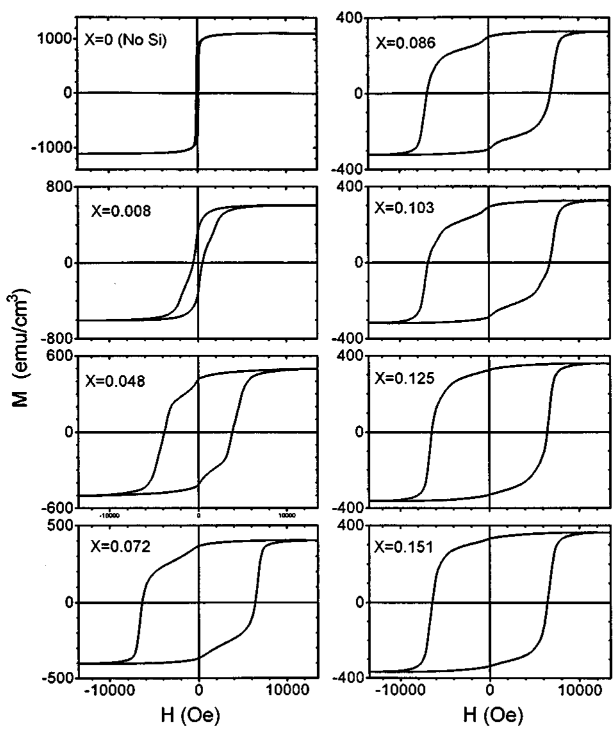

FIG. 1. Effects of Si content on magnetization hysteresis loops for $(Sm_2Fe_{12})_{1-x}Si_xC_y$ films.

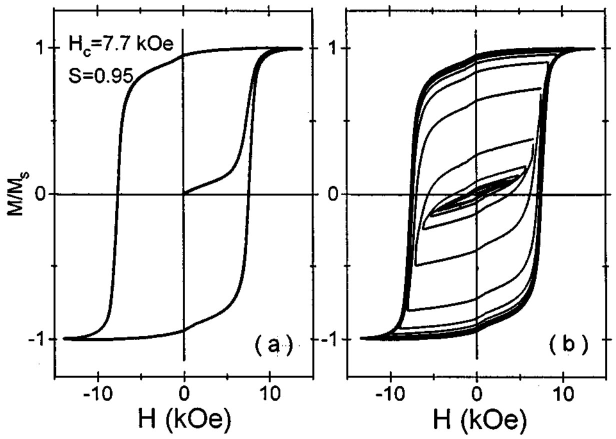

FIG. 2. Hysteresis loop and initial curve normalized by saturation magnetization (a) and minor loops (b) for the film with $x=0.125$.

coercivity film was never made without Si addition, even after very carefully adjusting the Ar pressure and annealing temperature.

Figure 2 shows typical initial curve and minor loops of our samples. Specifically this sample with $x=0.125$ shows coercivity of 7.7 kOe which is the highest H_c observed; this was obtained by carefully adjusting the preparation conditions. Both the initial curve and minor loops show a wall-pinning feature of reversal mechanism,[8] which will be discussed in more detail below. There is a small ''kink'' around the zero applied field in Fig. 2 and this kink becomes more appreciable for the films with narrower loops in Fig. 1. Possibly this feature originates from the existence of the soft phase which has larger content in films with smaller x values.

The effects of Si on coercivity H_c and saturation magnetization M_s are summarized in Figs. 3(a) and 3(b) respectively. It is interesting to see that in the range of $x=0$ to 0.086, H_c increases linearly with Si content x. Considering the wall-pinning feature of reversal mechanism as shown in Fig. 2, this linear relationship between H_c and x may imply that pinning centers were induced by the Si atomic sites in this x range. For $x>0.086$ in Fig. 3(a), H_c decreases slightly with increasing x. The magnetization M_s first decreases rapidly and then slowly with increasing the Si content x. Kronmüller *et al.*[9] have developed a model of magnetization reversal considering ''defect'' regions which have a different exchange constant and anisotropy from those of the host region. M_s decreases with x which suggests that Si atomic sites will show lower local magnetization and consequently different local exchange constant and anisotropy. Also, a high coercivity film was never made without Si addition. Thus the Si presumably affects the magnetic behavior by substituting for Fe but it also may introduce other defects such as grain boundary segregation, etc. This inference is consistent qualitatively with the experimental data as shown in Fig. 3(a), i.e., the linear relationship between H_c and x. However, the physical origin of the magnetization behavior is not clear and remains for future studies.

In nanocrystalline films, it is interesting to ask whether the structural grain and magnetic grain (or magnetic switching volume) have the identical size. Here we use the approach of the dependence of H_c on the applied field sweep rate[4,5] to determine the switching volume V^*. It has been shown[4,5] that the coercivity varies linearly with the logarithm of the field sweep rate, i.e.,

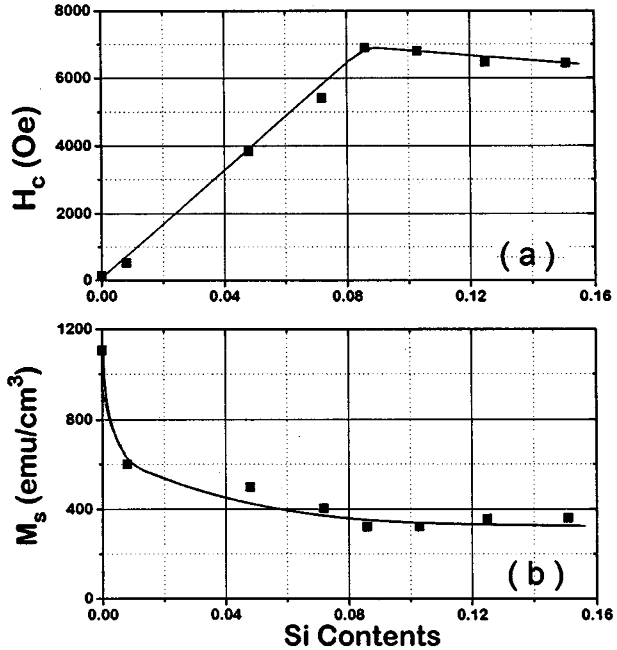

FIG. 3. Effects of Si content on coercivity H_c (a) and saturation magnetization M_s (b) for $(Sm_2Fe_{12})_{1-x}Si_xC_y$ films.

$$H_c = C + \frac{k_B T}{V^* M_s} \ln\left(\frac{dH}{dt}\right), \tag{1}$$

where C is a constant independent of (dH/dt), and k_B and T are the Boltzmann constant and temperature, respectively. Therefore the switching volume can be determined from the slope of the experimental curve of H_c vs $\ln(dH/dt)$.

Such measurements for all samples shown in Fig. 1 have been performed and two examples are shown in Fig. 4(a) for the film without Si addition and in Fig. 4(b) for the film with $x=0.125$. It is seen clearly that there are fairly good straight lines in Figs. 4(a) and 4(b) which imply that Eq. (1) is suitable for our films. The estimated V^* values from the slope of both straight lines are 2.1×10^{-17} cm^3 for the film without Si addition and 1.6×10^{-18} cm^3 for the film with Si content $x=0.125$ as listed in Figs. 4(a) and 4(b) as well. High resolution transmission electron microscopy indicates that the structural grain size for the film with $x=0.125$ is about 15 nm^2 which is consistent with the dimension of the magnetic grain size, i.e., $(V^*)^{1/3}=(1.6\times10^{-18}\ \text{cm}^3)^{1/3}\approx12$ nm. The measured V^* values for all films are summarized in Fig. 4(c). The significant effect of Si addition on V^* is seen: even for very small values of $x(0.008)$, V^* drops by a factor of $\sim$4.

Micromagnetism calculations[10,11] have shown that the coercivity behavior is closely related to the intergrain interactions. Also one may speculate that the above strong effect of Si addition on the switching volume V^* is correlated with

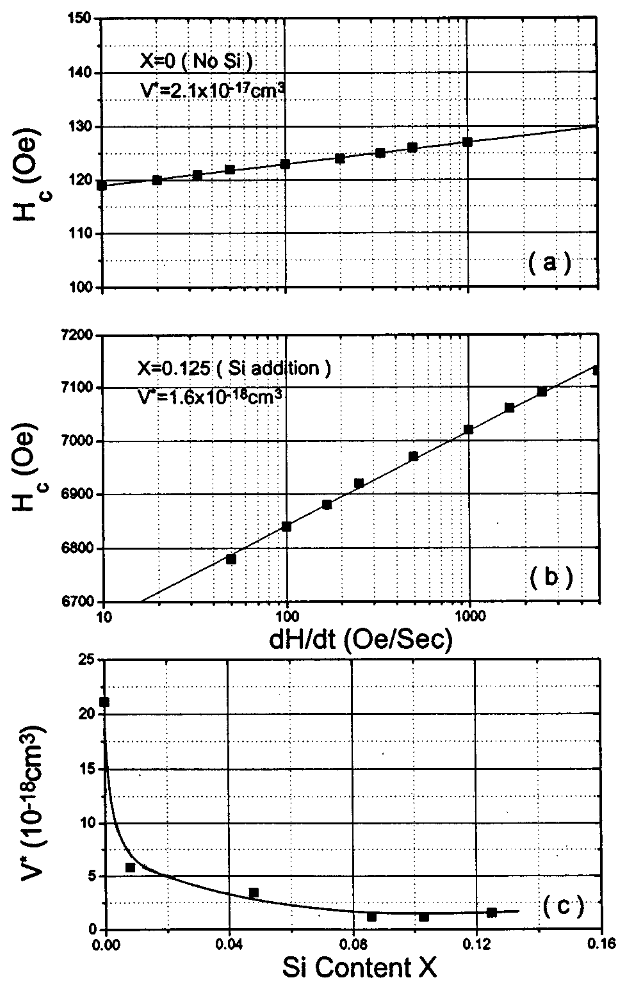

FIG. 4. Field sweep-rate dependence of H_c for the film with $x=0$ (a), $x=0.125$ (b), and the effect of Si content on magnetic switching volume V^* (c).

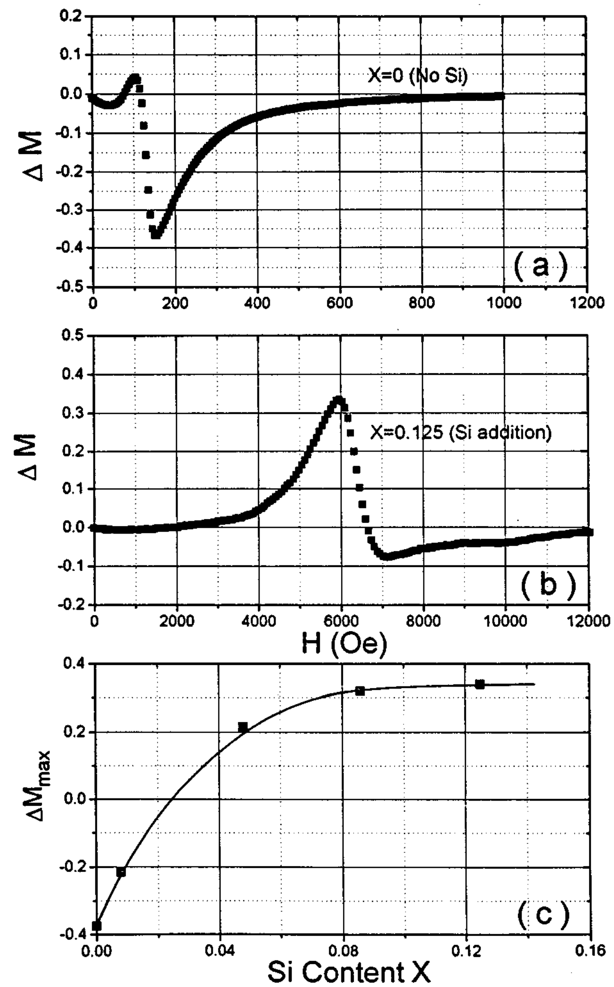

FIG. 5. ΔM curve for the film with $x=0$ (a), $x=0.125$ (b), and effect of Si content on the peak value of ΔM (i.e., $\Delta M_{\max}$) for $(Sm_2Fe_{12})_{1-x}Si_xC_y$ films.

the interactions between the grains. To investigate these interactions, the so-called ΔM method[3] has been used for selected films. Here ΔM is defined as

$$\Delta M(H)=I_d(H)-[1-2I_r(H)], \tag{2}$$

the exchange interaction dominates and negative ΔM means dipolar interaction dominates, and the amplitude of ΔM denotes the strength of the intergrain interaction.

Two examples of ΔM curves for $x=0$ and $x=0.125$ are shown in Figs. 5(a) and 5(b), respectively, and the peak value of ΔM, i.e., $\Delta M_{\max}$ as a function of Si content x are given in Fig. 5(c). We notice that the $\Delta M_{\max}$ curve changes from negative value to positive value. Normally one expects small values of $\Delta M_{\max}$ to indicate relatively small intergrain interactions and larger H_c values. In these films, however, small V^* values are correlated with large ferromagneticlike $\Delta M_{\max}$ values and high coercivities. These facts suggest the need for detailed studies of the nanostructure including possible intergranular phases. Such TEM work is underway.

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