Crystallographic Orientation Dependence of Sputtering Rate in Sendust Targets

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Abstract

The orientation dependence of sputtering rate in the sendust polycrystalline targets was studied. It was found from the present work that the erosion process is not uniform from one grain to another even within a target because of its polycrystalline nature showing many different orientations of grains. The grains oriented to promote efficient erosion were characterized by the close-packed planes which have large interplanar spacing and strong binding energy. The characteristic line patterns appeared on as-sputter target surface are discussed in terms of symmetry of crystallographic planes.

Keywords: Sputtering, Sendust, Crystallographic orientation, Atomic packing density, Polycrystalline, Symmetry, Binding energy, X-ray diffraction

1. Introduction

The deposition of alloys by sputtering is one of the most popular techniques for the preparation of alloy thin films because of its greater variety of materials and a good composition control. Basically, it comes from the fact that target atoms are dislodged from the target by momentum transfer from the impinging energetic ions. The higher momentum transfer to the substrate causes the film layers to grow with higher density, resulting in harder, smoother, and more stable films which are less susceptible to moisture absorption. In addition, the greater reactivity of the plasma environment promotes the formation of chemical bonding between film and substrate, leading to the greater adhesion. It has been shown that the microstructure of sputtering targets affect sputtering performance and film quality deposited. [1] The performance of sputtering targets is a function of the deposition process, the cathode design for a given sputtering system, and target microstructure. Previous studies on atom-ejection patterns governing the spatial distribution of sputtered atoms for various materials show that the most preferred atom-ejection direction in face-centered cubic materials is the <111> directions.[1, 2] However, only a little work has been done on the crystallographic orientation dependence of sputter yields in ordered polycrystalline materials. The main purpose of this work is to investigate the orientation dependence of sputtering rate in the sendust polycrystalline targets and to discuss its implication.

2. Material and Methods

The Fe-Al-Si sendust target showing the soft magnetic properties was used to study the crystallographic orientation dependence of sputtering rate in polycrystalline alloy targets. It has been known that this alloy is suitable for superior recording head core because of its large saturation flux density and high mechanical hardness.[4] The chemical composition of the specimen is summarized in the Table I. The condition of process variables during whole sputtering of the sendust target was fixed at 3 mTorr of Ar-gas pressure and 1.1 KW of RF power to minimize the dependence of process parameters during sputtering experiment. In addition, the experiment was done at the constant system geometry. For the microstructural examination as well as crystallo-graphic analysis of samples, test

specimens were cut out from the three different conditions of target surface. Those are as-received, as-sputtered, and as-lapped target samples. As-sputtered sample surface was also imaged under the optical and scanning electron microscopes (SEM) to study microstructural features on the surface of sputtered sendust target. In addition, the sputtered target surface was analyzed by means of X-ray diffraction technique (XRD, Rigaku @ 3KW with Cu K_α) for crystallographic information of planes exposed to plasma environment. Finally, microstructural features from analytical microscopy were tried to correlate to the crystallographic information from X-ray diffraction.

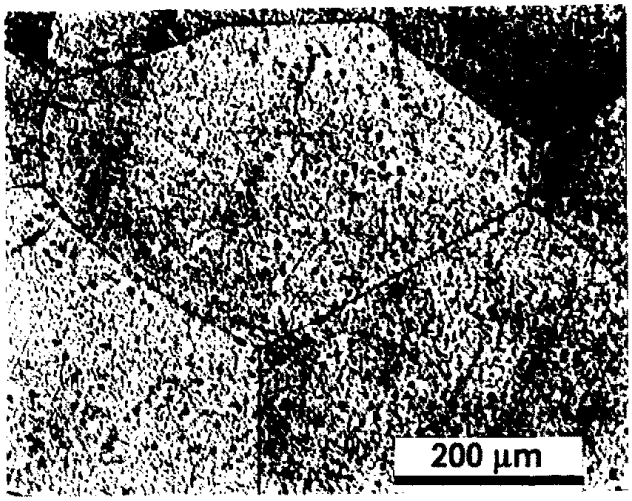

Figure 1.Optical micrograph of as-polished microstructure of the bulk sendust target

3. Experimental Results and Discussion

Figure 1 shows an optical micrograph of as-polished microstructure of the bulk sendust target used for the present study. As can be seen from the micrograph, the sendust target consisted of polyhedral shape of grains with an average grain size of about 270μm. The typical X-ray diffraction spectra from three different conditions of target surfaces (as-received, as-sputtered, and as-lapped surfaces of the target) are given in Figure 2. It is noted from the as-received sample (Fig. 2a) that some strong peaks including (220), (400), (200), and (111) planes are clearly observed. These results are comparable to the calculated values from $Fe_3Al(Si)$ powder sample summarized in the Table II except some deviation, possibly due to the small portion of preferred orientation in the as-received target. [3] In contrast to the Figure 2(a), the diffraction intensity of peaks from the as-sputtered target surface reveals totally different feature (Fig. 2b). In other words, the most strongest peak is from (224) plane rather than (220) plane which is reverse order in X-ray intensity table. Therefore, it is clear from these data that sputtering rates of different crystallographic planes are not the same. In addition, Table II(b) showing the measurement values of three samples indicates that the sputtering rate of the close-packed planes such as (220) in DO_3 superlattice is much faster than that of the loosely packed planes like (224). Figure 3 shows a cross-sectional view of the as-received target revealing "flat" surface before sputtering. In order to investigate the topographic variation of target surface during sputtering, the surface of as-sputtered target was imaged under an optical microscope. Typical examples of optical micrographs of as-sputtered target surface are given in Figure 4. As can be seen from the Figure 4(a), as-sputtered target surface shows the step-like feature of morphology revealing the different erosion rate.

Table I. The chemical composition of the bulk sendust target

Element	wt.%
Al	5.8
Si	10.1
Fe	balance

On the other hand, the X-ray spectra from the as-lapped sample after removing the step-like feature resulted from sputtering is quite similar to the that of as-received sample in trend. As suggested earlier, these results can be interpreted by considering sputtering mechanics in poly-crystalline targets and the binding energies for different crystallographic planes. Sputtering

consists of the acceleration of the heavy ions created in the plasma through an electric field and the impingement and removal of atoms from the target by mainly momentum transfer. In this situation, it is expected that the sputtering yield and the angular distribution of the sputtered fluxes are affected by the crystal structure of the target surface and its orientation. Wehner studied the non-uniform angular distribution of single crystals in detail and suggested that near threshold the sputtered atoms were ejected in the direction of close-packed atoms.[1] It is interesting that the angular dependence of the sputter yield for single crystalline target shows distinct peaks for ejection directions, related to be characterized by low Millier indices. Based on the Roosentaal's work, there was energy dependence of the sputter yields of Ar^+ on the (100) , (110) and (111) planes

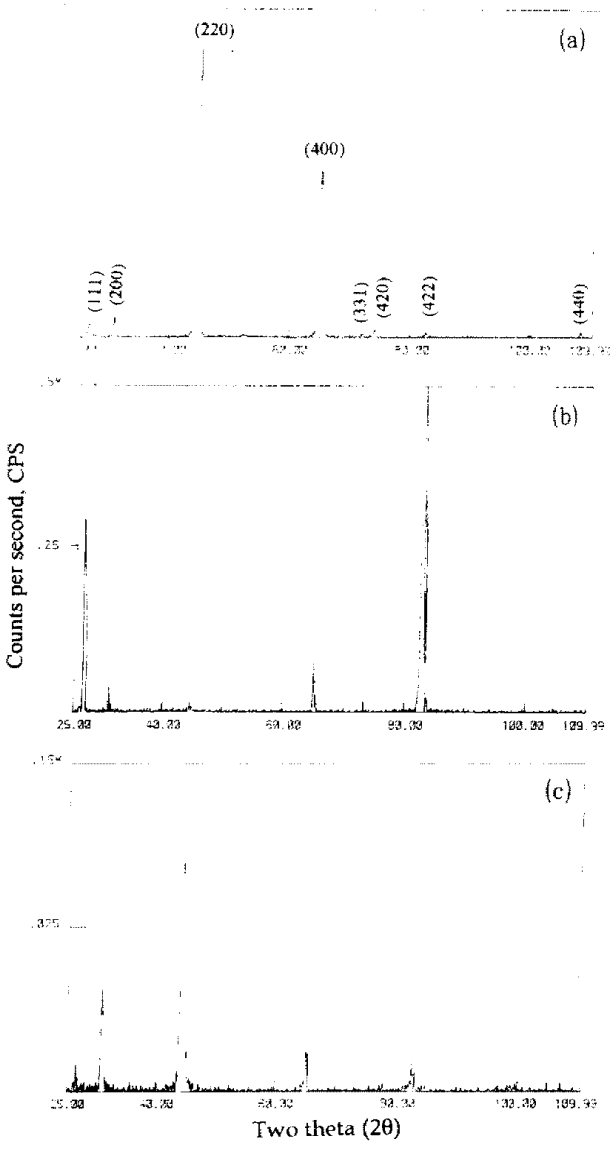

Figure 2. X-ray diffraction spectra from the samples (a) As-received, (b) As-sputtered, and (c) As-lapped

Table II. X-ray diffraction intensity of some planes in $Fe_3Al(Si)$ samples.

(a) Calculated values from the powder sample

(hkl)	$d_{(hkl)}$ (Å)	2θ (°)	I/I_o *
220	2.022	52.54	100.0
422	1.168	100.11	36.1
440	1.011	124.56	18.9
400	1.430	77.51	15.1
111	3.302	31.45	10.2*
200	2.860	36.48	5.2*
113	1.725	62.53	4.1*

* Normalized relative intensity
* Spectrum peaks from superlattice.

(b) Measurement values from the samples

As-received*1		As-sputtered*2		After lapping*3	
(hkl)	I/I_o	*(hkl)*	I/I_o	*(hkl)*	I/I_o
220	100	*224*	100	*220*	100
400	58.3	*111*	7.0	*200*	43.5
200	8.1	*400*	1.7	*400*	19.4
111	5.2	*200*	0.9	*111*	12.0
420	2.9	*220*	0.4	*224*	11.1
440	1.8				
224	1.8				

*1 From the as-received target surface
*2 From the as-sputtered target surface
*3 After lapping away steps on target surface

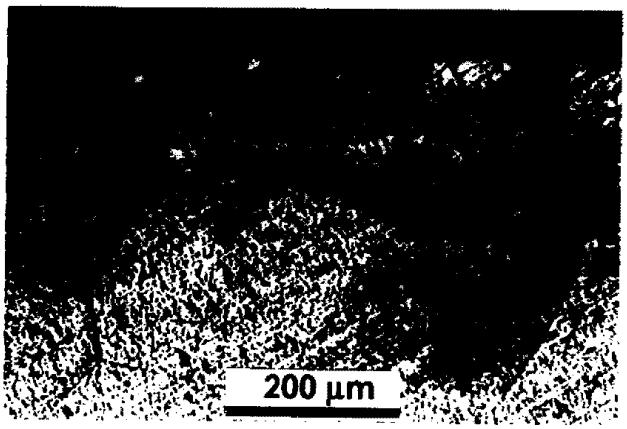

Figure 3. Cross-sectional view of the as-received target showing the intial flat surface

of face centered-cubic (FCC) material like copper.[4] In the range of 1~100 KeV of ion energy, the (111) plane showed the highest sputter rate (atoms/ion).

Another interesting feature from the Figure 4(b) is that the characteristic line patterns appeared for small portion of grains (arrowhead region). To get some ideas on the DO_3 superlattice, a schematic diagram showing the unit cell of DO_3 structure is shown in Figure 5. The sendust alloy can exist in a disordered state or ordered state.[5] Different kinds of atoms (Fe, Al or Si in this case) in disordered state will be distributed completely at random, while the atoms of each kind segregate to sites which form a regular pattern in an ordered state (superlattice). If the alloy is slowly cooled for target casting, the ordered state would be formed

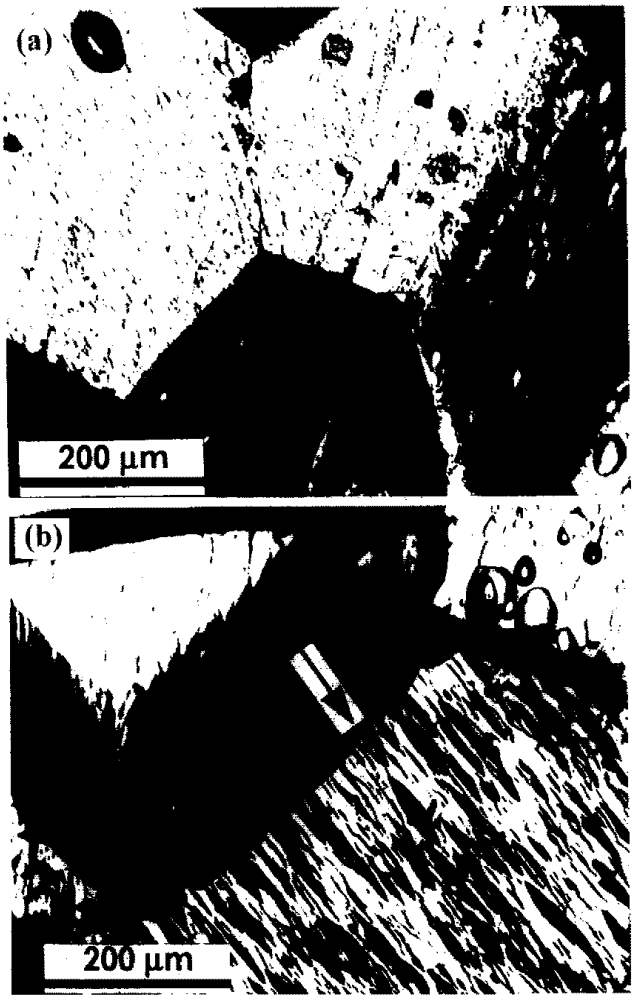

Figure 4. Optical micrograph of as-sputtered sendust target surface. (a) Step-like feature, and (b) Characteristic erosion patterns showing two-fold symmetry).

as temperature decreases. The characteristic line patterns appeared on as-sputtered sample root in two-fold symmetry on the (110) plane since there are two close-packed directions in the plane.

Then next question will be why the close-packed plane shows faster erosion rate. This led to speculation on the binding energy and sputter yield. When an energetic particle impinges on a solid and penetrates, it shares part of its energy with target atoms in a series of collisions. Fast recoils are generated which in turn set other target atoms in motion. A continuously increasing number of progressively slower atoms participate in the formed cascade. After the short period of time, transferable energies have become less than the energy needed to displace a further target atom. Finally, the cascade is damped by energy dissipation through phonon-assisted processes typically requiring ~10^{-10} sec. During the cascade development some of the target atoms in the near-surface region (smaller than 10Å in depth) acquire sufficient energy and momentum which is outwardly directed surmount the surface escape energy barrier and leave the target. In fact, there are several factors determining preferential sputter behavior. First of all, binding forces acting on an atom at the surface or in the bulk are species-dependent. In order to get sputtered, an atom must first be set in motion during the process of energy dissipation by primary radiation. Finally, atoms may be ejected not only from the top surface layer of a solid or liquid but also from a shallow depth range underneath. Jackson calculated surface binding energies of some cubic metals as a key quantity determining the energies of surface ejection processes by using Morse potentials.[6] The well-known Morse potentials due to Girifalco and Weizer is given by

$$\varphi_M = D\,[\exp\{-2\alpha(r-r_o)\} - 2\exp\{-\alpha(r-r_o)\}] \quad \ldots\ldots (1)$$

These potentials are summed over ideally perfect lattices. The surface binding energy, U_1 was defined as the potential energy between an atom in the surface and the rest of the crystal. It is the minimum energy required to remove an atom

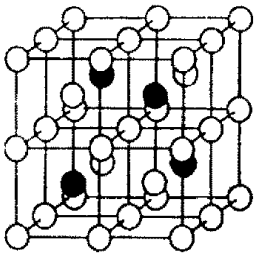

Figure 5. Schematic drawing of the ordered DO_3 structure in $Fe_3Al(Si)$ alloy

from the surface and take it to infinity. Similarly U_2 refers to the layer below the surface and so on for the U_N's.

$$U_1 = I(0) + \Sigma I^*(z) \quad \text{..... (2)}$$

U_1 consists of two contributions which are $I(0)$ and $I^*(z)$. In the above expression, $I(0)$ is the potential energy due to all the atoms in the surface plane while the second term represents the potential due to the rest of the underlying semi-infinite solid. Approximately, it came up with following values of U_1 for different planes of cubic metals. As can be seen from the table, the binding energy for a given plane in BCC metals which are quite similar to DO_3 in atom arrangement is inversely proportional to the planar atomic packing density. Therefore, the grains oriented to promote efficient erosion were characterized by their close-packed planes being bombarded. and a wide interplanar spacing between them.

Table III. Atomic packing density (APD) and the estimated values of U_1's for different planes in cubic metals

	APD (100)*1	APD (110)	APD (111)
FCC	78.4%	55.5%	90.6%
BCC	58.9%	83.4%	33.5%
	U_1 (100)	U_1 (110)	U_1 (111)
FCC	1.31 U_o*2	1.20 U_o	1.33 U_o
BCC	1.30 U_o	1.34 U_o	1.11 U_o

*1 A hard sphere model assumed.
*2 U_o: the cohesive energy of a crystal (commonly taken as a sublimation energy in eV/atom)

4. Conclusions

Based on the present work, the following conclusions can be drawn. The sendust alloy target showed non-uniform erosion pattern in a microscopic scale during sputtering even within a target. As a result, a step-like feature on the as-sputtered target surface was obtained. This was originated from the different rate of removal of material from different planes because of its polycrystalline nature. The grains oriented to promote efficient erosion were characterized by their close-packed planes being bombarded. The difference in erosion rate is due to the different atomic packing density which is also related to the binding energy. It was also noted from the microstructural analysis that the characteristic line lorphology appeared on the as-sputtered surface and its origin root in two-fold symmetry on the (220) plane in the sendust alloy target.

5. References

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