Effect of Excimer Laser Annealing on Crystallization and Atomic Ordering of $Co_2Mn_{0.5}Fe_{0.5}Ge$ Heusler Alloy Thin Films toward Spintronic Applications

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Heusler alloys exhibit intriguing material properties, including high spin polarization, large anomalous Hall effect and low damping, making them highly attractive for spintronic applications. One major challenge in the fabrication of Heusler-alloy-based thin film devices is high-temperature treatment required to promote the atomic ordering to obtain the full potential, which is often incompatible with the temperature restriction of the practical device fabrication. In this study, we explore excimer laser annealing (ELA) as an alternative annealing method to address the above temperature constraint. By conducting ELA right after the deposition of Heusler-alloy-based film, steep temperature gradient along the depth direction can allow the film to reach sufficient temperature, keeping the underlying structures within the temperature limitation. We apply ELA to $Co_2Mn_{0.5}Fe_{0.5}Ge$ (CMFG) Heusler alloy thin films and found that crystallization and atomic ordering of the CMFG films were induced similar to the conventional furnace annealing, without interdiffusion or roughness increase. These results demonstrate the potential of ELA as a viable annealing technique, thereby expanding the application of Heusler alloys.

Index Terms—**Excimer laser annealing, Heusler alloys, Spintronics, Magnetic recording**

I. Introduction

Heusler alloys exhibit unique properties such as high spin polarization, large anomalous Hall effect and low damping. These properties are expected to greatly enhance the device performance in spintronic application. In current-perpendicular-to-plane giant magnetoresistance (CPP-GMR) read heads [1], and anomalous Hall effect read heads [2], studied for next-generation hard-disk-drives (HDDs), high spin polarization and large anomalous Hall effect are crucial for enhancing the output. Low damping can improve the efficiency of magnetization oscillation and magnetization switching in microwave-assisted writing devices [3,4] and magnetoresistive-random-access-memory (MRAM) cells. To fully realize the potential of the Heusler alloys, heat treatment is usually conducted to promote atomic ordering. However, device fabrication in practical applications imposes temperature limitations. In the case of HDD read heads, process temperature is limited to 300°C due to the temperature tolerance of the soft magnetic shield layers. In the case of MRAM, the process temperature of memory cells fabricated in the back-end-of-line process is limited to 400°C.

Here, we explore excimer laser annealing (ELA) [5] as an alternative annealing method for Heusler alloys to address the above conflicting temperature requirements. By applying a pulse laser after the deposition of a Heusler-alloy-based device film, steep temperature gradient along the depth direction is induced as laser absorption and subsequent heat generation occurs at the surface. This temperature gradient can allow the film near the surface to reach the required temperature, while keeping the underlying structures within the temperature limitation. Although ELA has already found large-scale practical use such as in fabrication of poly-crystalline Si on glass or flexible substrates, where amorphous Si layers are rapidly heated to the melting point for crystallization before heat diffusion into the substrate causes damage, its application to spintronic Heusler alloys remains unexplored.

II. Experimental

The samples consisting of Ta(2 nm) / CoFeBTa(1.5 nm) / CMFG(50 nm) / Ru(2 nm) were prepared by sputter deposition on thermally oxidized Si substrates. The composition of CMFG was estimated by X-ray fluorescence (XRF) to be $Co_{1.992}Mn_{0.712}Fe_{0.372}Ge_{0.928}$. Pulsed KrF excimer laser with a wavelength of 248 nm, a pulse duration of 83 ns was introduced to the homogenizing optics to form uniform rectangular fluence profile and applied to the sample surface. The irradiated area was 0.369 mm × 1.198 mm. A series of 1000 laser pulses was applied at a repetition rate of 10 Hz. Crystallization and atomic ordering of the samples were examined using X-ray diffraction (XRD). Surface morphology was measured using atomic force microscopy (AFM). Anisotropic magnetoresistance (AMR) and resistivity measurements were conducted by applying a current of 1 mA to the strips with a width of 40 µm and length of 170 µm and measure the resistance by the four-terminal method.

III. Results and Discussions

Figure 1(a) shows the photo image of the ELA sample for various fluence levels from 40 to 300 mJ/cm^2. The position marks, denoted by the red box, were made by damaging the film with high fluence laser, and the 2 mm wide area next to the position guide, denoted by the blue box, is the ELA area made by sweeping the laser spot. Noticeable degradation of Surface specularity occurred when the fluence is 160 mJ/cm^2 or more, suggesting a change in surface morphology. Figures 1(b) shows the corresponding out-of-plane XRD profiles. For fluence below 100 mJ/cm^2, the profiles show no peaks, indicating the amorphous structure. However, from 100 mJ/cm^2, 004, 220 fundamental peaks, and 002 superlattice peaks appear, demonstrating that the crystallization and *B*2 atomic ordering of the CMFG layers were successfully induced by ELA. At 200 mJ/cm^2, the peaks become sharp and intense, presumably due to growth of larger grains by agglomeration.

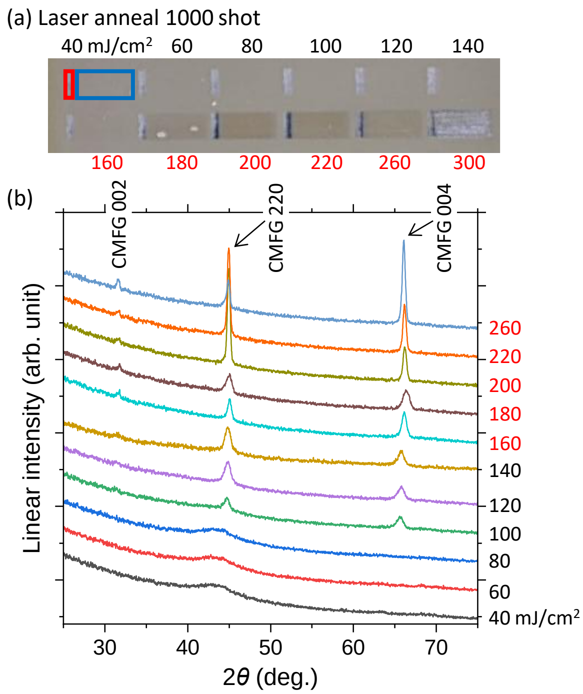

FIG.1. (a) Photo image of the ELA sample. (b) Out-of-plane XRD profiles obtained for the ELA samples for various fluence values.

Figure 2 shows the fluence dependence of average roughness (R_a). In the as-deposited state, the sample surface is very flat with R_a of approximately 0.15 nm. The roughness remains unchanged up to 140 mJ/cm^2 and starts to increase monotonically with fluence. Such increased roughness due to excess fluence is detrimental in device fabrication in practical applications. The ELA conditions with increased roughness shows overall agreement with the specularity degradation.

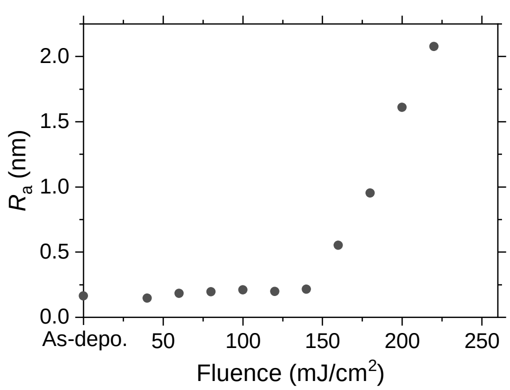

FIG. 2. R_a of the as-deposited and ELA samples as a function of fluence.

Figure 3 shows resistivity and AMR ratio as a function of fluence. The resistivity decreased from 120 mJ/cm^2 and shows a large drop at 140 mJ/cm^2 followed by continuous decrease to a minimum at 180 mJ/cm^2. This resistivity change is attributed to the improved atomic ordering and growth of the crystal. Beyond this fluence, the resistivity shows an upturn, presumably reflecting the increased roughness and interdiffusion due to excessive fluence. The AMR ratio is almost zero in the as-deposited state, and becomes negative after ELA, showing similar trend to that of the resistivity. The negative AMR is associated with the half-metallicity as discussed in Ref. [6]. Notably, at fluence of 140 mJ/cm^2, crystallization and atomic ordering of CMFG with relatively large negative AMR was obtained with no roughness increase, revealing the optimal ELA condition and demonstrating the applicability of ELA to the fabrication of Heusler-alloy-based spintronic devices.

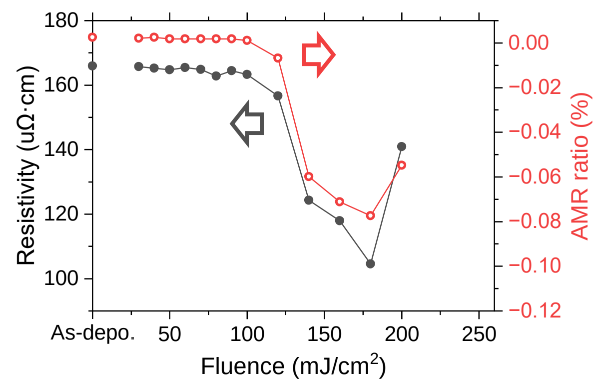

FIG. 3. Resistivity and AMR of the as-deposited and ELA samples as a function of fluence.

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