Thermal Stability of Co-Fe and Cu Multilayered Thin Films


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Intermixing in sputter deposited Co-Fe and Cu thin film multilayers is both of scientific interest and technological importance. Due to their near complete immiscibility in bulk form, the formation of intermixed multilayers seemingly presents a thermodynamic paradox. Furthermore, due to the use of Co-Fe and Cu thin films in the magnetic storage industry, there is interest in associating the degradation in giant magnetoresistive sensor performance with atomic migration in these layers. We have studied Co-Fe (Co with a few atomic percent Fe) and Cu multilayers, each layer of 2.5 nm thickness, in as-deposited and several annealed conditions with TEM, X-ray diffraction, differential scanning calorimetry (DSC), and three dimensional atom probe (3DAP).

The as-deposited condition, Fig. 1(a), shows Co-Fe and Cu layers of relatively high purity. A 28 day anneal at 310 °C, Fig. 1(b), shows drastic changes in multilayer morphology, where a large cluster of Cu has grown to the point at which it now crosses at least three Co-Fe layers. The multilayer structure is still intact to the right, where it is observed that the Cu layers are thinner due to the migration of Cu atoms toward the growing Cu particle. This multilayer breakdown is in accord with thermodynamic predictions that suggest the multilayer structure forces significant contact between Co-Fe and Cu at the interfaces, whose free energy can be reduced by breaking up into a two-phase mixture of Co-Fe and Cu particles. Monte Carlo simulations predict that the breakup of Co/Cu multilayers into a two-phase structure is initiated by grain boundary grooving [1].

However, after annealing at 410 °C for only 5 minutes, Fig. 1(c), a partially intermixed state is observed (coexisting with the clustering at grain boundaries), which appears to be in stark contrast to the immiscibility shown in bulk Co-Cu and Fe-Cu phase diagrams. DSC results also show evidence of the intermixing in the form of an endothermic heat pulse around 400 °C. Furthermore, theoretical thin film calculations, which include interfacial and strain energy, suggest that at 400 °C the components of the overall free energy which contribute to intermixing are approximately equal in magnitude to the components which contribute to phase separation. Therefore, it may be possible for the system to evoke a lattice diffusion mechanism at higher temperatures, in addition to the grain boundary mechanism active at lower temperatures, which is fueled by entropy and the heat release provided by the destruction of interface and lattice strain. X-ray
diffraction results show that this intermixed state is only temporary as the grain boundary clustering mechanism eventually predominates, forming the equilibrium two-phase mixture.

![2-D cross-section cuts through atom probe datasets. (a) as-deposited. (b) after 310 °C, 28 day anneal (shown in lower magnification). (c) after 410 °C, 5 min anneal. Co-Fe in blue, Cu in red.]

FIG. 1. 2-D cross-section cuts through atom probe datasets. (a) as-deposited. (b) after 310 °C, 28 day anneal (shown in lower magnification). (c) after 410 °C, 5 min anneal. Co-Fe in blue, Cu in red.

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