

Enhancing Exchange Bias with Diluted Antiferromagnets

Jung-Il Hong,¹ Titus Leo,² David J. Smith,² and Ami E. Berkowitz^{1,3}

¹Center for Magnetic Recording Research, University of California–San Diego, La Jolla, California 92093, USA

²Department of Physics and Astronomy and Center for Solid State Science, Arizona State University, Tempe, Arizona 85287, USA

³Department of Physics, University of California–San Diego, La Jolla, California 92093, USA

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The exchange bias H_E of coupled polycrystalline films of antiferromagnetic CoO and ferromagnetic Co was significantly enhanced by the systematic substitution of nonmagnetic Mg for Co in CoO. Samples in which either Co or $\text{Co}_{1-x}\text{Mg}_x\text{O}$ were deposited first were investigated at temperatures from 10 to 300 K. With $\text{Co}_{1-x}\text{Mg}_x\text{O}$ on the bottom, the increased interfacial uncompensated spin density of the single antiferromagnetic domain $\text{Co}_{1-x}\text{Mg}_x\text{O}$ crystallites produced the enhanced H_E . With Co on the bottom, a thin interfacial oxide layer was primarily responsible for the strongly increased H_E .

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The shifted hysteresis loop, or exchange-bias field H_E resulting from exchange anisotropy [1], in bilayers of ferromagnetic (FM) and antiferromagnetic (AFM) layers has been studied extensively [2,3]. It is generally accepted that H_E is related to the coupling between the FM layer and uncompensated interfacial spins in the AFM layer, with the density of interfacial uncompensated AFM spins determining the magnitude of the exchange field [3]. This suggests that an enhancement of H_E could be achieved by increasing the interfacial AFM uncompensated spin density. H_E enhancement has been demonstrated in bilayers with AFM CoO on Co, in which some Co atoms in the top CoO layer were replaced with nonmagnetic Mg atoms, or by adding excess oxygen in the form of Co_3O_4 phases in the top CoO [4]. The epitaxial (CoMg)O films were (111) textured, with grain sizes 25–35 nm and roughness ~ 6 nm. A 0.4-nm interfacial layer of CoO was inserted between the FM and AFM layers. These bilayers exhibited maximum H_E at $\sim 10\%$ Mg and a monotonic decrease of H_E with temperature increase for all Mg concentrations. The results were explained with a “domain state” model, in which domains formed in the AFM, with the domain walls being stabilized by the presence of nonmagnetic Mg ions within the AFM layers [4,5].

In our present work, polycrystalline bilayer films were grown with either $\text{Co}_{1-x}\text{Mg}_x\text{O}$ or Co deposited first. The widths of the randomly oriented individual $\text{Co}_{1-x}\text{Mg}_x\text{O}$ grains were so small (< 10 nm) that each grain was a single AFM domain, with strong magnetocrystalline anisotropy at low temperatures. With $\text{Co}_{1-x}\text{Mg}_x\text{O}$ on the bottom, H_E was increased by the increased density of uncompensated interfacial AFM spins resulting from the Mg substitution, with the most effective Mg substitution being consistent with numerical modeling. This increase augmented the uncompensated spin density arising from finite size effects in the AFM grains [6]. With Co on the bottom, H_E was enhanced by a thin discontinuous interfacial CoO layer, with high uncompensated spin densities, that formed during deposition of the $\text{Co}_{1-x}\text{Mg}_x\text{O}$.

The bilayers were deposited at room temperature by dc magnetron sputtering onto Si (100) substrates with their native oxide. Two sets of bilayers (sets 1 and 2) were deposited. In both sets, oxide layers with thicknesses of 20 nm were deposited by reactive sputtering from metallic Co and Mg targets in a mixed Ar and O_2 atmosphere. The MgO deposition rate was controlled relative to the CoO deposition rate in order to vary the Mg concentration x in the $\text{Co}_{1-x}\text{Mg}_x\text{O}$ AFM layers. For set 1, the $\text{Co}_{1-x}\text{Mg}_x\text{O}$ was deposited first and a 6-nm Co film was deposited on top of it, followed by deposition of a 5-nm-thick SiO_2 capping layer. For set 2, the 6-nm Co layer was deposited first, with the $\text{Co}_{1-x}\text{Mg}_x\text{O}$ layer deposited on top. Hysteresis loops were measured in a SQUID magnetometer after the bilayers were field cooled from 350 to 10 K in a field of 2.5 T (the Néel temperature of $\text{Co}_{1-x}\text{Mg}_x\text{O}$ is ≤ 293 K). To remove any training effect, the samples were field cycled 20 times between ± 2 T before the hysteresis loops were recorded.

All AFM oxide films were polycrystalline and roughly columnar, as shown in the plan-view [Fig. 1(a)] and cross-sectional view [Fig. 1(b)] transmission electron micrographs of samples from set 1. Average grain widths ranged from ~ 9 to ~ 6 nm as the fractional Mg concentration was increased. Nanoprobe energy-dispersive x-ray spectroscopy of the CoMgO layers showed that the Mg atoms were distributed homogeneously within the oxide layer, without significant evidence for second phase formation or segregation to grain boundaries.

Figure 2 summarizes the exchange-bias fields for set 1 ($\text{Co}_{1-x}\text{Mg}_x\text{O}$ on bottom), at temperatures from 10 to 300 K. H_E at 10 K increases from 230 to 361 Oe as x increases from 0 to 0.14, and then decreases for $x \geq 0.14$, becoming 0 for $x > 0.65$. The temperature dependence of H_E as a function of x is plotted in Fig. 2(b). H_E exhibits a plateau before beginning to decrease at temperatures close to the blocking temperature T_B . This trend is similar to the results of Takano *et al.* [6] but different from the behavior reported by Keller *et al.* [7] for the AFM on top. The plateau becomes less flat for $x > 0.14$.

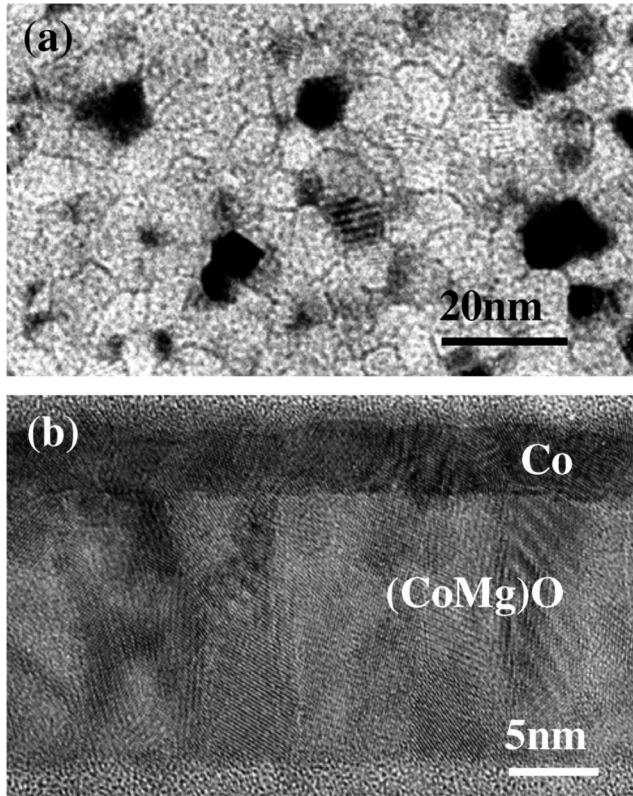


FIG. 1. Transmission electron micrographs showing typical size and morphology of (CoMg)O samples from set 1: (a) plan view of $\text{Co}_{0.88}\text{Mg}_{0.12}\text{O}$, (b) cross-sectional view of a Co/ $\text{Co}_{0.85}\text{Mg}_{0.15}\text{O}$ bilayer.

The increase of H_E for x between 0 and 0.14 can be understood in terms of an increase of the uncompensated spin density in the AFM oxide layer that results from replacing Co with Mg. As nonmagnetic Mg atoms begin to replace Co atoms, some spins of neighboring Co atoms become uncompensated, and the uncompensated spin density is roughly proportional to the density of Mg atoms. However, as x is further increased, the additional Mg atoms become less efficient at creating uncompensated spins. The average distance between Mg atoms becomes smaller, and spins of Co atoms around the newly added Mg atoms may have already become uncompensated due to other nearby Mg atoms. Clusters of Mg atoms will also begin to form, and uncompensated Co spins exist around these clusters. However, the number of uncompensated spins created for each added Mg atom is then smaller than when all Mg atoms remain separated. As x increases even further, Mg atoms replace Co atoms with uncompensated spins, without producing additional uncompensated spins; this results in a decrease of uncompensated spin density.

We have calculated the uncompensated spin density as a function of Mg concentration based on the AFM oxide structure. In CoO, the Co atoms form a face-centered-cubic structure with an oxygen atom between every pair of Co atoms. The first and second nearest neighbors of each Co

atom are exchange coupled via oxygen atoms. The Mg dilution was simulated by progressively replacing randomly chosen Co atoms with Mg atoms. At selected Mg concentrations, the spins of Co atoms that were first and second nearest neighbors of each substituted Mg atom were counted as uncompensated. The number of Co atoms with uncompensated spins on specific crystallographic planes was then counted. The fraction of uncompensated spins for the (111) plane is plotted in Fig. 2(c) as a function of Mg concentration. The maximum uncompensated spin density occurs for $x \approx 0.15$ for the crystallographic planes considered, i.e., (100), (111), and (110). This value is close to the measured value of 0.14 for which H_E at 10 K is maximum, as shown in Fig. 2(a). For $x > 0.14$, the calculated uncompensated spin density decreases, as discussed above. Furthermore, the blocking temperature T_B and the anisotropy of $\text{Co}_{1-x}\text{Mg}_x\text{O}$ also decrease as x increases, producing the rapid decrease of H_E observed for $x > 0.14$. At higher temperatures, the maximum H_E occurs at progressively lower x because T_B decreases with increasing x . The temperature dependence of H_E below the blocking temperature may also involve the formation of Mg clusters. Uncompensated spins around Mg clusters are likely to be more temperature-dependent, because their coupling to the bulk AFM structure is weaker due to the higher concentration of nonmagnetic Mg neighbors. We note that the increased uncompensated spin density due to the Mg dilution increases H_E above the values established by the finite size effects in the $\text{Co}_{1-x}\text{Mg}_x\text{O}$ grains [6].

The values H_E for set 2 (Co layer on bottom) are plotted in Fig. 3. H_E from 10 to 50 K exhibits a broad maximum as a function of x , with very little decrease at 100% Mg. At higher temperatures, the maximum H_E moves to smaller x , as observed for set 1 in Fig. 2(a). However, the general dependence of H_E on both x [Fig. 3(a)] and temperature [Fig. 3(b)] was clearly different from those of set 1. In particular, it is noteworthy that H_E is not suppressed even at $x = 1$, which nominally corresponds to Co/MgO bilayers. All samples exhibited a monotonic decrease of H_E as the temperature increased, and all bilayers with $x > 0.40$ have the same T_B of 175 K.

To explain this behavior, we note that the Co layers in set 2 were briefly exposed to O_2 when it was introduced into the sputtering chamber prior to deposition of the (CoMg)O layers by reactive sputtering. Therefore, a thin discontinuous layer of CoO oxide forms on top of the Co film, as sketched in Fig. 3(c). Cross-sectional electron micrographs of the set 2 bilayers, as shown, for example, in Fig. 3(d), confirmed that the AFM-FM interfaces were rougher and less abrupt than for the set 1 bilayers, consistent with the formation of a discontinuous native oxide [8]. The existence of this interfacial CoO is confirmed by the high values of H_E when $x = 1$ (nominally pure MgO). The interfacial oxide consists of a mixture of CoO and (CoMg)O, with an average composition containing less

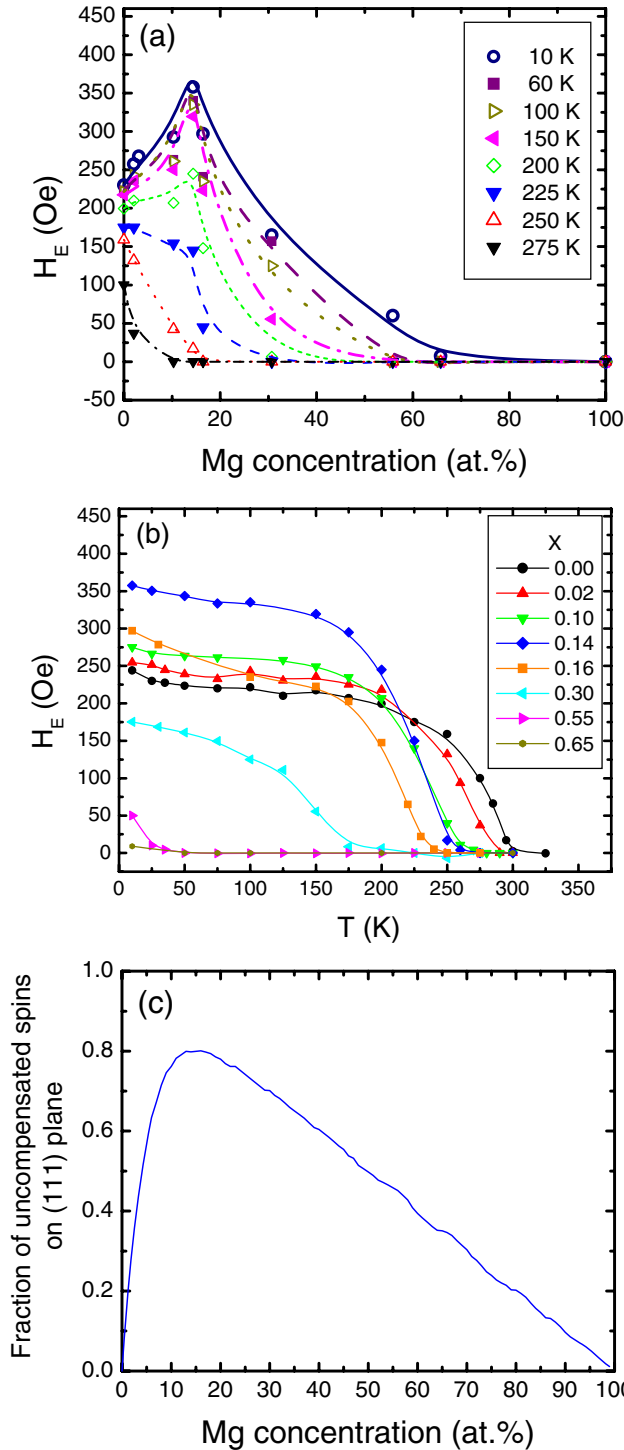


FIG. 2 (color online). H_E for set 1 ($\text{Co}_{1-x}\text{Mg}_x\text{O}$ underneath): (a) as a function of Mg concentration x at selected temperatures and (b) as a function of temperature for samples with selected x . (c) Calculated uncompensated spin density on (111) planes.

Mg than deposited and with considerable roughness, which would increase the uncompensated spin density [2,3]. At $x = 0$, the roughness is smoothed out, and H_E is minimum. As x increases, the CoO nanoparticles become more dis-

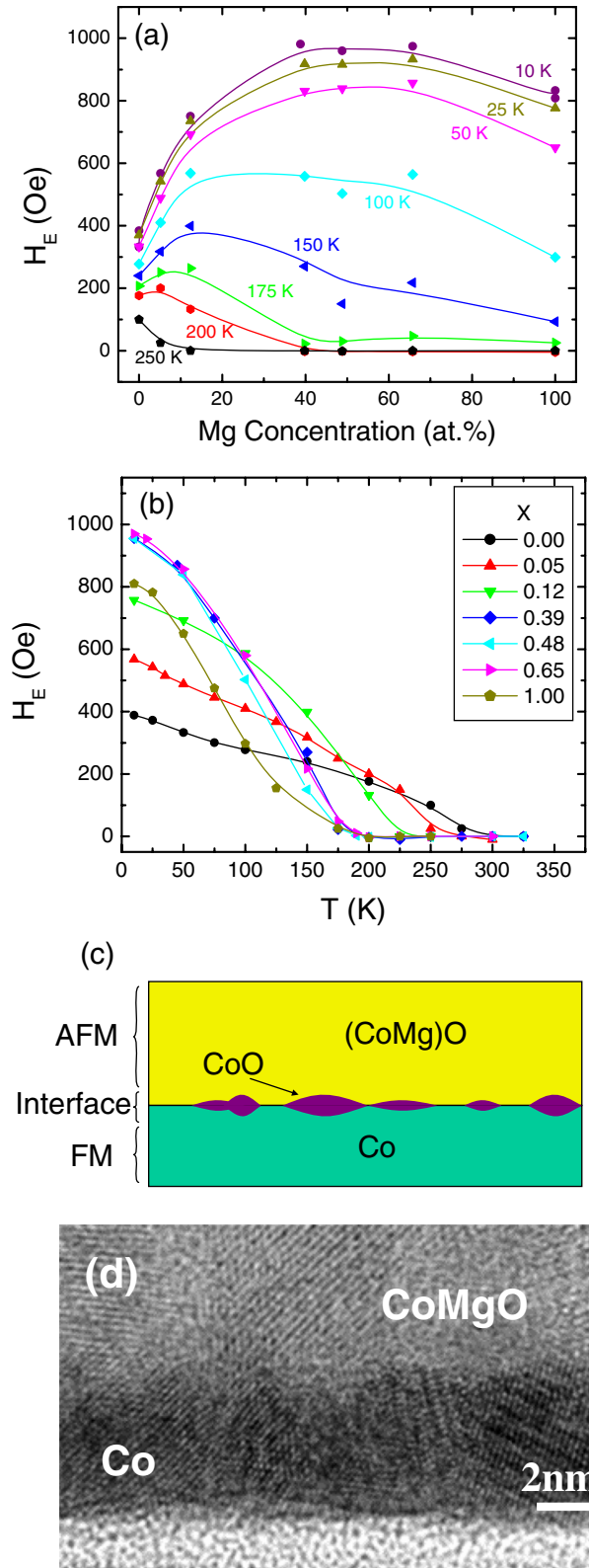


FIG. 3 (color online). H_E for set 2 (Co layers underneath): (a) as a function of Mg concentration x at selected temperatures and (b) as a function of temperature for selected x . (c) Sketch of interface structures with native oxides. (d) Cross-sectional electron micrograph showing a region of a Co/ $\text{Co}_{0.61}\text{Mg}_{0.39}\text{O}$ bilayer.

crete, and their high uncompensated spin densities are increasingly effective. For $x > 0.4$, the high H_E is relatively constant, suggesting that it is dominated by the exchange coupling between the discontinuous CoO and the Co metallic layer. Another contribution to higher H_E is the fact that oxidation of the Co layer reduces its thickness, and H_E is generally regarded as being inversely proportional to FM thickness [2,3]. Exchange coupling between a Co film and its native oxide has been studied previously [9–12]. A monotonic decrease of H_E with temperature as well as a T_B of ~ 175 K were reported, similar to the behavior for $x > 0.4$ in Fig. 3(b). For $x < 0.4$, H_E increases and T_B decreases with increasing Mg concentration.

In summary, this investigation has shown that the exchange field in polycrystalline Co/Co_{1-x}Mg_xO bilayers can be enhanced significantly by substituting nonmagnetic Mg atoms into the AFM layers for either Co or Co_{1-x}Mg_xO deposited first. For Co_{1-x}Mg_xO on the bottom, the dependence of H_E enhancement on Mg dilution was correctly predicted by an atomic-scale numerical model. This enhancement augments the uncompensated spin densities produced by finite size considerations in the single domain Co_{1-x}Mg_xO grains. For Co on the bottom, H_E is primarily due to the discontinuous CoO native oxide that is produced when oxygen is introduced in the sputtering chamber for the reactive deposition of the Co_{1-x}Mg_xO. The observation that H_E is barely reduced when only MgO is deposited supports this conclusion.

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