Nanoscale manipulation of thin film magnetic properties can be achieved through proximity effects or exchange coupling in bilayer or multilayer structures. The close contact of the layers enables direct insight into the fundamental problems of magnetic and electronic correlations and can lead to structures and functionalities with engineered control for technological applications. For example, the proximity of a ferromagnetic film with a vanadium oxide (VO₂ or V₂O₃) results in a reversible modification of the film’s magnetic coercivity due to interfacial stress. This is caused by the magnetoelastic anisotropy adding to the thin film coercivity due to interfacial stress. The origin of the interfacial stress is the Structural Phase Transition (SPT) in vanadium oxides. The SPT coincides with the Metal Insulator Transition (MIT), which is discontinuous and phase coexistence of metallic and insulating states can be observed.

In this paper, we present the effect of nanoscale phase coexistence during the MIT of V₂O₃ on the magnetic properties of a ferromagnetic Ni thin film. We observe a maximum coercivity peak that coincides with the V₂O₃ phase transition on top of an overall increase of the coercivity with decreasing temperature. We propose that this sharp increase arises from a length scale competition between magnetic domains of Ni and phase coexistence during the V₂O₃ phase transition. This model is supported by micromagnetic simulations and shows that magnetic properties of ferromagnetic films are strongly affected by a proximal first order phase transition. © 2014 AIP Publishing LLC.

Coercivity enhancement in V₂O₃/Ni bilayers driven by nanoscale phase coexistence

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We studied the temperature dependence of coercivity and magnetization of V₂O₃/Ni bilayers across the Structural Phase Transition in V₂O₃. We found a coercivity peak that coincides with the V₂O₃ phase transition. Micromagnetic simulations were performed with the Object Oriented Micromagnetic Framework (OOMMF) package from NIST. The narrow temperature region in which the maximum change in the magnetic properties is observed may be useful for technological applications. The effects are fully reversible, persistent at each temperature, and show a thermal hysteretic behavior.

V₂O₃/Ni bilayers were prepared in a high-vacuum sputter deposition system with base pressure of 1 × 10⁻⁷ Torr. 100 nm V₂O₃ films were deposited onto (1012) sapphire substrates at 750 °C in 4 mTorr of ultra-high purity (UHP) Ar by RF sputtering at 100 W of a V₂O₃ target. 10 nm thick Ni layers were deposited after cooling to room temperature and recovering the base pressure. A 5 nm capping layer of Nb was deposited to prevent oxidization. Ni and Nb were deposited in 4 mTorr of UHP Ar by RF sputtering at 100 W. Temperature dependent structural characterizations were carried out using a Rigaku Smartlab X-ray diffractometer operating between 80 K and 300 K. Magnetization as a function of field and temperature was recorded using a Quantum Design PPMS-DynaCool Vibrating Sample Magnetometer (VSM) and a Quantum Design superconducting quantum interference device (SQUID) magnetometer, respectively. Electrical transport measurements were performed in a two-probe configuration using a constant current source. The morphology of the thin films at room temperature was investigated with an atomic force microscope (AFM) operating in tapping mode. Since the structural and electronic configuration of the V₂O₃ film crucially depends on the temperature, a slow sweep rate of 1–2 K/min was used in all measurements. Thus, thermal lagging was minimized and reproducible measurements were obtained throughout the V₂O₃ phase transition. Micromagnetic simulations were performed with the Object Oriented Micromagnetic Framework (OOMMF) package from NIST.

Fig. 1(a) shows the magnetization as a function of field for selected temperatures throughout the V₂O₃ phase transition. The data have been normalized by the Ni volume and shows symmetric ferromagnetic hysteresis with varying
values at 200 K. The values of the coercivity below 150 K and above 185 K are independent of the temperature sweep direction. Fig. 1(c) shows the behavior of the magnetization during heating of the sample with a 500 Oe applied magnetic field. A minimum in the magnetization occurs at the same temperature as a maximum in the $H_C$. The reduced magnetization is observable in Fig. 1(a), with the hysteresis loop at 165 K having the lowest magnetization at 500 Oe. The remnant magnetization is correlated with the coercivity and increases (decreases) as the loop width increases (decreases).

The changes in $H_C$ and magnetization coincide in temperature with the $V_2O_3$ phase transition, Fig. 2. The MIT observed in the resistance measurements as a function of temperature is shown in Fig. 2(a). Typical $V_2O_3$ thin films show up to seven orders of magnitude change during the MIT. The resistance values are dominated mostly by the metallic Ni film, the Nb capping layer, and the metallic contact resistance. This leads to the linear behavior and a suppression of the absolute change resulting from the MIT, Fig. 2(a). Nevertheless, a distinct change in resistance is observed between 150 K and 175 K with a thermal hysteresis typical for the $V_2O_3$ first order transition.

The SPT accompanying the MIT is shown in Fig. 2(b). Each point on the curves corresponds to an individual out-of-plane x-ray diffraction measurement at a constant temperature. Above 185 K a single Bragg peak is observed at $2\theta_b = 24.35^\circ$, corresponding to the (012) plane of the rhombohedral phase of $V_2O_3$. During the SPT, the high temperature rhombohedral phase transforms into the low temperature monoclinic phase. In the diffraction measurements, this manifests in a Bragg peak observed at $2\theta_M = 24.07^\circ$, which can be identified with the (110) monoclinic lattice direction. The correct indexing of the high and low temperature diffraction peaks has been confirmed by

width. The coercive fields $H_C$ as a function of temperature are summarized in Fig. 1(b). During cooling of the sample, a maximum of $H_C = 211$ Oe was observed at 160 K. This maximum is shifted by about 5 K to higher temperatures for the measurements during heating. This shift is consistent with the thermal hysteresis observed in the $V_2O_3$ first order transition. The maximum is asymmetric and the values at lower temperatures remain over a factor of two higher than the
measurements along different directions with respect to the surface normal. At each temperature, the data are fitted assuming two Gaussian functions centered at 2\theta_K and 2\theta M, respectively. These Gaussian fits are integrated and normalized to the highest values obtained for each phase. The measurement is repeated for the heating and cooling temperature branch, leading to the four intensities shown in Fig. 2(b). Both MIT and SPT show a 5 K thermal hysteresis in the 180–150 K range. The transition temperatures in the 100 nm films are similar to those observed in bulk V_2O_3. The centers of MIT and SPT are located at the same temperatures at which the maximum coercivity and minimum magnetization are observed.

Increased coercivity below 150 K compared to the values above 190 K was previously observed and attributed to stress in the Ni film. Above 190 K, the Ni film is in the as-grown relaxed condition and shows lower H_C values. Below 150 K, V_2O_3 has undergone the structural phase transition and the Ni grown on the top is stressed, thus showing higher H_C values. This highlights the direct coupling between the V_2O_3 and Ni films. However, this explanation alone does not account for the maximum of the coercivity observed in the middle of the V_2O_3 phase transition, Fig. 1(b). Additional experiments have shown that a change in coercivity is still present when the V_2O_3 layer is decoupled from the Ni film by a thin Cu spacer layer (data not shown). This indicates that the antiferromagnetic ordering in V_2O_3, if present, is not playing a crucial role in the coercivity enhancement. Furthermore, we have performed measurements in field cooling configurations and all magnetic hysteresis were reproduced without any indication of exchange bias.

We developed a model considering the phase coexistence throughout the first order phase transition and the length scales involved in the coercivity enhancement. We consider crystallographic grains of Ni and V_2O_3 to be of the same size and larger than the lateral domain size of Ni. The crystallographic grain size and the intrinsic Ni magnetic domain size can be considered temperature independent. However, in V_2O_3 both metallic and insulating phases coexist across the transition with varying lateral extension.

Well above the V_2O_3 transition, all magnetic domains in the Ni volume are strain-relaxed and the film has a defined value of coercivity. With decreasing temperature, insulating monoclinic domains nucleate in V_2O_3. Parts of the Ni film, which are in contact with these domains, show a higher coercivity due to stress. The boundary between stressed and unstressed Ni can be considered as a localized structural and magnetic defect. Upon magnetization reversal, this defect acts as a pinning center for magnetic domain wall movement and thus further enhances the coercivity. As the V_2O_3 phase transition progresses, the Ni film becomes more inhomogeneous and reaches a maximum disorder in the center of the transition at which both phases coexist equally, Fig. 2(b). The maximum disorder in the center of the transition produces a maximum in the coercivity, Fig. 1(b). In addition to the increased coercivity, the presence of additional domains reduces the Ni magnetization. This decrease in magnetization within the phase transition is consistent with the experimental observations in Fig. 1(c). Further, cooling of the sample shifts the V_2O_3 phase concentration towards the insulating state and larger monoclinic domains coalesce. As a consequence, the absolute number of pinning sites in the Ni decreases and the coercivity reduces. The final value of the coercivity well below the phase transition temperature corresponds to the stressed Ni film without extra pinning. Coercivity enhancement due to a related phenomenon was observed in STO/Nd_0.5Sr_0.5CoO_3 films. In this case, the coercivity enhancement is due to an interfacial phase separation leading to additional pinning centers.

The lateral extension of magnetic domains and the domain of the SPT is crucial for observation of this behavior. A different lateral disorder, for example, caused by a higher roughness, reduces the magnetic domain size and increases the number of domain walls. Therefore, the initial value of the coercivity above 190 K is expected to be higher. Further, the Ni domains are smaller than the V_2O_3 domains and less additional domain walls are created by the V_2O_3 SPT. Therefore, no peak is observed in the coercivity in the middle of the transition. This behavior can be observed comparing a V_2O_3/Ni bilayer with 3 nm interface roughness with a sample that has a lower roughness of 1.5 nm, Fig. 3. More importantly, the sample with lower roughness showed terraces of 100 nm width on which the roughness remains below 1 nm, Fig. 3(b). Samples with higher roughness do not show this lateral correlation. For the case of lower roughness, the high temperature values for the coercivity are reduced by more than a factor of two and a clear maximum enhancement is observed, Fig 3(d).

To investigate the hypothesis that nanoscale phase coexistence in V_2O_3 causes magnetic domain separation and domain wall pinning in Ni, we performed a micromagnetic simulation using OOMMF. We model the Ni film with a 1 \mu m x 1 \mu m x 10 nm Ni slab with an exchange constant of A = 3.4 J/m and saturation magnetization M_S = 494 kA/m. The slab was discretized into a mesh of 5 x 5 x 10 nm^3 cells. For the simulations, we assumed that the Ni and V_2O_3 grains have the same size and the Ni slab is divided into 100 nm x 100 nm grains according to our AFM measurements, Fig. 3(b). Based on the results shown in Fig. 2, we assumed that the transition temperatures for individual V_2O_3 grains have a mean value of 165 K and follow a Gaussian distribution with a standard deviation of 5 K. Ni domains in contact with a V_2O_3 domain which has undergone the transition are attributed with a randomly oriented anisotropy of K = 1 x 10^3 J/m^2. The random orientation of the anisotropy is justified considering that Ni is textured only along the out-of-plane direction, but is likely to show polycrystallinity in the film plane. This anisotropy breaks the Ni film into ~100 nm domains during the magnetization reversal. At the boundary between monoclinic and rhombohedral phases of V_2O_3 we introduce an additional anisotropy due to lattice mismatch of differently strained Ni grains. This lattice mismatch provides the additional domain wall pinning and has been parametrized with an anisotropy of K = 4 x 10^4 J/m^3. The width of the boundary is optimized to 30 nm in order to reproduce Fig. 1(b) quantitatively. Note that the domain wall width parameter (\delta \approx (A/K)^{1/2}) is 18 nm for K = 1 x 10^4 J/m^3 and A = 3.4 pl/m, implying that the Ni domain wall width expected from theory is similar to the width of the boundary chosen here. The simulated values of coercivity
and magnetization as a function of temperature are shown in Fig. 4. At 165 K, a maximum pinning is observed due to largest amount of boundaries. The simulated coercivity reaches a maximum of 180 Oe and the magnetization reaches a minimum, caused by the formation of the magnetic domains. As the cooling continues, all V$_2$O$_3$ grains switch to the monoclinic structure, and therefore the boundary becomes smaller and disappears when the phase transition in V$_2$O$_3$ ends. In the final state, the phase boundaries and therefore the pinning sites in the Ni film are removed. Only by adjusting the magnitude of the anisotropies and the boundary width, a quantitative agreement between experiment (Fig. 1) and simulation is achieved (Fig. 4). Our simulation does not take into account the general temperature behavior of the Ni film coercivity and magnetization with temperature and thus does not reproduce the evolution of H$_C$ above or below the transition.

In summary, nanoscale phase coexistence of a first order phase transition strongly affects the magnetic properties of a ferromagnetic layer that is in contact. Inhomogeneities and phase coexistence across the V$_2$O$_3$ SPT are responsible for a large enhancement in the coercivity and decrease in the magnetization. Both effects occur over the narrow temperature window of the phase transition. A micromagnetic simulation based on magnetic domain wall pinning at phase boundaries agrees with the experimental data. The reversibility and large tunability of the effect make it feasible for technological applications in which nanoscale manipulation is required.

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17The anisotropy values and the width of the boundary have been chosen to obtain good agreement between simulations and experiment.