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Coupling of ferromagnetism and structural phase transition in V$_2$O$_3$/Co bilayers

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Abstract

Interfacial coupling in hybrid magnetic heterostructures is being considered as a unique opportunity for functional material design. Here, we present the temperature dependence of magnetic properties of V$_2$O$_3$/Co bilayers influenced by the structural phase transition that is accompanied by a metal–insulator transition in V$_2$O$_3$. Both the coercivity and the magnetization of Co layer are strongly affected by the interfacial stress due to the magnetostrictive effect in the ferromagnetic film. The observed change in coercivity is as large as 59% in a narrow temperature range. The changes in the magnetic properties are reproducible and reversible, which are of importance for potential applications.

Keywords: metal–insulator transition, magnetostrictive coupling, structural phase transition, heterostructure

(Some figures may appear in colour only in the online journal)

1. Introduction

Manipulating magnetic properties of ferromagnetic (FM) layers through stimuli other than a magnetic field has attracted a lot of research attention in recent years due to its potential for device applications [1–3]. Examples of external stimuli include electric field, electric current, pressure and light, which open new routes to tune the properties of magnetic materials in a controlled way [3–8]. Thin-film hybrid structures are particularly sensitive to external stimuli and they are ideal platforms to investigate the underlying physical mechanisms dominating the properties of magnetoelectric heterostructures, such as ferromagnetic/multiferroic, piezoelectric/ferromagnetic and ferroelectric/ferromagnetic heterostructures [9–11]. One promising approach that can exploit the manipulation of the magnetic properties is provided by placing a ferromagnet in proximity to materials that undergo a structural phase transition (SPT) accompanied by a metal–insulator transition (MIT) [5, 12–15]. In this case, the MIT and SPT can be induced not only by changing temperature, but also can be driven by pressure, light or electric current [5]. Therefore, these materials offer the possibility to tune the magnetic properties of the ferromagnetic layer by multiple stimuli.

Among all the materials that show a MIT and SPT, vanadium sesquioxide (V$_2$O$_3$) is one of the most fascinating systems due to its rich phase diagram [16, 17]. V$_2$O$_3$ shows a first-order structural phase transition (at ~160 K) from a high temperature metallic state to a low-temperature insulating state [18]. The change in resistance can be as large as 7 orders of magnitude across the MIT [5]. These remarkable features are associated with a SPT from a rhombohedral crystal structure in the metallic phase to a monoclinic crystal structure in the insulating phase that occurs simultaneously with the electronic phase transition [13]. The crystallographic change occurring in V$_2$O$_3$ provides
the possibility to achieve reversible manipulation of the magnetic properties of the FM layer in hybrid thin-film through the magnetoelastic anisotropy that is induced by phase coexistence at the nanoscale in the MIT material and the interfacial stress within the transition temperature range. As already reported in literature [12, 13, 15], the proximity of FM layers, such as Fe, Co or Ni, to V2O3 and VO2 affects the magnetic and electronic properties at the interface between FM and SPT materials. Hence, combining V2O3 film with a ferromagnetic Co film in a bilayer configuration provides a unique way to investigate the coupling effect when the oxide undergoes a SPT accompanied by a MIT. However, the coupling effect between V2O3 and Co has not been studied in detail.

In this paper, we describe the fabrication of V2O3/Co bilayers grown by molecular beam epitaxy (MBE) and investigate their temperature-dependent magnetic properties. We find that the ferromagnetic properties of the Co thin film in the bilayers are strongly affected by interfacial stress associated with the SPT accompanied by a MIT in V2O3. Reversible changes are observed for the coercivity and magnetization across the SPT in the V2O3 film. This effect results in a reproducible modification of magnetic properties, which is of direct relevance for technological applications.

2. Experimental details

The V2O3/Co bilayers are grown by MBE in a chamber with a base pressure of $1 \times 10^{-9}$ mbar. 30 nm-thick V2O3 films are deposited on (0001)-Al2O3 substrates at a temperature of 742 °C and in an oxygen partial pressure of $8.3 \times 10^{-7}$ mbar. 5 nm-thick Co films are then ex situ deposited on the V2O3 thin films as well as on bare Al2O3 substrates at room temperature.
temperature and at a base pressure of $5 \times 10^{-9}$ mbar [19]. To prevent oxidization, 3 nm of Au is deposited in situ as capping layer on top of the Co layer. The thin-film samples are characterized by means of high resolution x-ray diffraction (XRD) using Cu Kα radiation. The surface morphology of the Co film on V₂O₃ is characterized by in situ scanning tunneling microscopy (STM) before capping with Au. The temperature-dependence of the resistance of the V₂O₃ film is measured using a four-probe geometry and a constant current source. Temperature is swept at a slow rate of 1–2 K min⁻¹ with a Lakeshore 332 temperature controller. Magnetic properties are measured with a superconducting quantum interference device magnetic property measurement system (SQUID-MPMS, Quantum Design).

3. Results and discussion

In figure 1(a) we present the XRD scan for a 30 nm-thick V₂O₃ film grown on an (0001) oriented Al₂O₃ substrate. It is obvious that the sample shows good crystalline quality and the absence of any secondary phase. In this case, only the peak corresponding to the (0006) plane of V₂O₃ is observed, indicating that the V₂O₃ film grows epitaxially on the substrate along the (0001) orientation. To determine the microscopic structure of the Co film, we present in figure 1(b) the STM image of a 5 nm-thick Co layer on V₂O₃. Nearly circular, coalescing Co grains are observed with typical size smaller than 8 nm and with a RMS roughness of 0.52 nm. The inset of figure 1(b) shows the height profile across the line indicated in the 2D map. The RSM roughness and height profile confirm the successful preparation of the V₂O₃/Co bilayers, which allows performing a reliable investigation of the heterostructural magnetic properties.

To investigate the coupling between V₂O₃ and Co, the magnetization curves of the V₂O₃/Co bilayers are measured at different temperatures near the SPT in the V₂O₃. The results are plotted in figure 2(a). All curves show a ferromagnetic behavior with varying coercivity, originating from the top Co layer. Figure 2(b) summarizes the dependence of the coercive field $H_c$ on temperature for the V₂O₃/Co bilayer compared to a reference Co film grown directly on Al₂O₃. For the Co reference film, we observe the standard (almost) linear dependence of the coercivity on temperature. For the bilayers, however, a strong deviation from the linear behavior of $H_c$ occurs within a narrow temperature range (160 K–185 K). For temperatures higher than 185 K, $H_c$ of the bilayer is almost the same as of the Co layer. However, an increase in $H_c$ of ~59% (from 92 Oe at 185 K to 146 Oe at 160 K) is observed in the bilayer while the change in the same temperature range for the Co film is of only ~15%. Interestingly, the deviation in $H_c$ is observed within a temperature range where the V₂O₃ structural phase transition occurs (in this temperature range the V₂O₃ layer exhibits a MIT, see figure 3(a)) [13, 20]. On the other hand, the coercive field in the V₂O₃/Co bilayers is slightly larger for increasing than for decreasing temperature (see inset in figure 2(b)), which can be attributed to the typical thermal hysteresis observed for the structural phase transition in V₂O₃ [12]. However, when the temperature is below 160 K and above 185 K, i.e. outside of the phase transition region, the coercivity shows a quasi-linear dependence on temperature and it is independent of the temperature sweep direction. We therefore can conclude that the observed anomalous ferromagnetism in the thin Co films is associated with the SPT in the adjacent V₂O₃ thin film.

In figure 3(a) we present the resistivity as a function of temperature for a V₂O₃ film. Inset shows the differential curves of resistivity versus temperature to indicate the transition temperature of 176 K. (b) Coercivity versus temperature and (c) ZFC magnetization measured at 50 Oe as a function of temperature for a V₂O₃/Co bilayer.
within the same temperature range where the deviation from the linear behavior of the coercivity is observed. Hence, the MIT and SPT are centered approximately at the same temperature at which the behavior of the coercivity and magnetization with temperature deviate from the Co reference film. Thus, the magnetic properties of the Co layer can be modified by relying on the SPT and MIT in the adjacent V$_2$O$_3$ layer. Note that the transition in resistivity typically spans a broad region in figure 3(a). This is because the transition in resistivity with decreasing temperature goes through four successive stages: a homogenous metallic state, a striped nanotexture of percolating electronic phase coexistence, an inhomogeneous correlated insulator state and a homogeneous insulator state [18]. As described in [18], in the region of coexistence of insulating and metallic domains below the bulk $T_{MIT}$, the transport is influenced by percolation.

In figure 4 the zero-field-cooled (ZFC) and field-cooled (FC) magnetization curves of a Co thin film at 100 Oe and of the V$_2$O$_3$/Co bilayer at different magnetic fields are shown. Since the contribution of the V$_2$O$_3$ to magnetization is negligible, all curves have been normalized with respect to the Co volume. From figure 4(a) it is clear that the Co thin film shows splitting of the ZFC and FC, which is likely due to the small size of Co grains contained in the film. The magnetization reveals a superparamagnetic behavior with 'blocking' occurring at a temperature higher than 350 K [22]. On the other hand, it is noticeable that there is a different behavior in the magnetization of the bilayers as already shown in figure 3(c) (see figure 4(b)). The magnetization (both in the ZFC and FC curves) shows a small kink in the same temperature range where the deviations from the linear behavior in the coercivity are observed, as shown in figure 3(b). This further indicates that both effects are closely related to the SPT in the V$_2$O$_3$ thin film. In figures 4(b)−(d) the splitting between the ZFC and FC magnetization at different magnetic fields disappears at higher temperatures, which can be attributed to a superparamagnetic behavior resulting from the presence of small Co grains (see figure 1(b)). We note that the ZFC magnetization gradually increases (grains become unblocked) until reaching the average blocking temperature $T_B$, while the FC curves slowly decrease with increasing temperature. Moreover, it is evident that $T_B$ tends to shift to lower temperatures when the applied magnetic field increases. This is expected and can be accounted for the fact that a larger magnetic field tends to reduce the energy barriers that hinder the fluctuation of magnetic moment, resulting in a transition to a thermally unfrozen state at lower temperatures.

As indicated above, another interesting finding is that the kink in magnetization, which is observed within the temperature interval where the SPT in the V$_2$O$_3$ thin film occurs, gradually vanishes when increasing the applied magnetic field, as illustrated in figures 3(c) and 4. Moreover, when the applied magnetic field is relatively low, these striking changes in the magnetization as function of temperature indicate that the Co
magnetization may be not fully saturated. On the other hand, the SPT in V$_2$O$_3$ appears to play a crucial role to induce these changes in the magnetic behavior of the V$_2$O$_3$/Co bilayers. It is well known that the magnetism of magnetic metal atoms is susceptible to an environment with fewer nearest neighbor atoms [23]. For example, surface atoms can reveal a strong magnetic polarization, while the bulk atoms have a smaller magnetic moment. Hence, the weaker interatomic hybridization at the V$_2$O$_3$/Co interface is likely to influence the magnetization of Co atoms at the interface. The symmetry and coordination number at the interface of the bilayers can be changed due to the SPT in the V$_2$O$_3$ film, which induces a narrowed d-band and therefore localizes surface states or surface resonance states in the Co atoms. This then results in a significantly stronger magnetic polarization in the interface region and can induce an anomaly in the magnetization for small applied magnetic fields [23, 24]. When the applied magnetic field is, however, increased to 500 Oe, the results in figure 4(d) illustrate that the coupling effect at the interface is suppressed and the magnetization of the Co layer is not affected by the SPT in the V$_2$O$_3$ layer. This indicates that the effect of weakened interatomic hybridization near the V$_2$O$_3$/Co interface becomes suppressed by the external magnetic field at which the Co may also be fully saturated.

As we mentioned above, the remarkably reproducible modification of the coercivity is also a result of the SPT in the V$_2$O$_3$ layer, which is accompanied by a MIT. The V$_2$O$_3$ SPT gives rise to a 1.4% volume increase when undergoing a transition from a rhombohedral phase at higher temperature to a monoclinic phase at lower temperature [21]. The volume expansion leads to epitaxial stress in the adjacent FM Co layer, thus changing the magnetic properties by an inverse magnetostrictive effect. The magnitude of the stress that produces the variations in coercivity shown in figure 2(b) can be estimated from the stress anisotropy field $H_{K\sigma}$, which is given by [25]:

$$H_{K\sigma} = \frac{3\lambda_{si}\sigma}{M_S}$$

where $\lambda_{si}$ is the saturation magnetostriction, $\sigma$ is the stress in MPa, and $M_S$ is the saturation magnetization in G or emu cm$^{-3}$. The observed value of coercivity at 120K, at which the SPT is expected to be completed, is 147 Oe and 201 Oe in the single Co film and the bilayer, respectively (see figure 2(b)). The additional anisotropy coercivity produced by the coupling effect is then 54 Oe. We calculate $M_S$ to be 632 emu cm$^{-3}$ according to the results presented in figure 2(a). However, there are large variations of reported $\lambda_{si}$ values for Co, ranging from $\sim -11 \times 10^{-6}$ to $-95 \times 10^{-6}$ [25–28]. Also, the Co layer is likely to be a collection of coalesced nanoparticles forming a continuous film. We therefore cannot make a good estimate of the stress produced by the V$_2$O$_3$ layer according to the equation (1), nor can we make a comparison with the coupling of a VO$_2$/Ni layer, which produces a $\sigma = 589$ MPa across the SPT [12]. However, we expect a stronger coupling effect when the Co films are grown in situ on top of the V$_2$O$_3$ films.

4. Conclusion

We observe magnetostrictive coupling across the interfaces of hybrid magnetic heterostructures. The coupling is caused by the interfacial stress induced by the structural phase transition accompanied by the metal–insulator transition in the V$_2$O$_3$ film, which causes pronounced changes in the magnetic properties of V$_2$O$_3$/Co bilayers due to the magnetostrictive effect. The change in coercivity is as large as 59% within a narrow temperature range across the transition. The reversible modification of the magnetic properties of ferromagnets opens up new possibilities for technological applications in which relevant ferromagnetic properties, such as magnetization and coercivity, can be tailored for desired applications.

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