

Coupled Phenomena

Control of the Magnetic Configuration of Ferromagnetic Nanostructures Across the Structural Phase Transition of Vanadium Dioxide

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Received 14 Jul 2016, revised 11 Aug 2016, accepted 30 Aug 2016, published 30 Sep 2016, current version 23 Nov 2016.

Abstract—We investigate the effect of the structural phase transition in VO₂ in magnetoelastically coupled heterostructures of VO₂ and Ni. Continuous and nano-patterned Ni layers were used, and we found reversible and reproducible magnetic domain switching induced by the VO₂ structural phase transition. The magnetic states of the nano-patterned ferromagnetic elements were dominated by topographic features which generated strong pinning but still allowed for a reversible switching between the states. Our measurements constitute a key step for the use of the VO₂ phase transition for ultrafast dynamical studies of the inverse magnetostrictive effect, and eventually employing the effect for ultrafast low-power switching devices.

Index Terms—Coupled phenomena, magneto-elastic materials, magnetic microscopy, multiferroics.

I. INTRODUCTION

Control of magnetic switching using an external driving force such as an electric field or strain is critical for the development of novel memory devices. This has led to active research on both natural [Fiebig 2005, Spaldin 2005] and artificial multiferroics [Spaldin 2005]. The most promising artificial multiferroics consist of magnetoelastically coupled heterostructures combining a ferromagnetic (e.g., Ni) and a piezoelectric (e.g., [Pb(Mg_{0.33}Nb_{0.66})O₃]_(1-x)-[PbTiO₃]_x) material. When an electric field is applied across the piezoelectric material, the resulting strain generates a magnetic anisotropy in the ferromagnetic material by the inverse magnetostrictive effect [Buzzi 2013, Cui 2015, Finizio 2013, Finizio 2014, Parkes 2014, Weiler 2009].

To assess the dynamic behavior of the inverse magnetostrictive effect, it is necessary to generate a strain variation on a timescale comparable with the magnetization dynamics (i.e., on the order of 10⁻⁹ s). The investigation of the dynamic properties of the inverse magnetostrictive effect has been, up to now, carried out by employing surface acoustic waves to generate the dynamically varying strain [Gowtham 2015, Weiler 2011]. However, only a periodic strain variation is easily achievable using surface acoustic waves. Possible alternative approaches to surface acoustic waves to generate fast variations of strain are to use a piezoelectric crystal in the linear regime, or to employ materials such as VO₂, which exhibits a metal-insulator transition (MIT) associated with a structural phase transition (SPT) from a monoclinic to a rutile phase on sub-ns timescales [Cavalleri 2001, Wen 2013]. This SPT can also be induced using heating, providing an easy-to-access experimental method to excite the VO₂ SPT; however, this comes at the price of being a slower process dominated by thermal diffusion. Such a SPT leads, for bulk VO₂, to relative strain variations of up to $\epsilon \approx 10^{-2}$ along the *c* axis of the rutile phase [Cao 2010]. Furthermore, because the phase transition in VO₂ is hysteretic,

this leads to bi-stable magnetic states, as required for non-volatile memory applications.

It was recently demonstrated [de la Venta 2013] that the magnetic properties of continuous magnetostrictive layers grown on top of VO₂ thin films are affected by crossing the SPT of the VO₂. However, as of now, no analysis of the influence of the VO₂ SPT on the magnetization configuration of magnetostrictive elements at the micro- and nanoscale has been carried out.

In this letter, we show that the magnetic state of nanostructured magnetostrictive elements fabricated on top of a thin VO₂ film can be manipulated by crossing the MIT of the VO₂. We study the reversibility of the magnetic state upon heating and cooling of the VO₂ across the SPT threshold temperature, to probe whether this interesting material can be employed to excite the magnetization dynamics in a pump-probe measurement, and thus be used for ultra-fast switching of devices.

II. EXPERIMENTAL

VO₂ films 100 nm thick were fabricated from a stoichiometric target by RF sputtering onto 0.5 mm thick *r*-plane Al₂O₃ (10–12) substrates, according to the parameters given in de la Venta [2013], growing in a textured phase with (110) out-of-plane orientation, and were subsequently capped with 2 nm of Al to preserve the oxygen stoichiometry of the as-deposited films. The Al-capped VO₂ films were analyzed by X-ray diffraction to verify the stoichiometry and the crystallinity, yielding results comparable to the ones reported in de la Venta [2013].

The resistance-vs.-temperature curves of the deposited VO₂ films were characterized by two point resistance measurements, as detailed in de la Venta [2013].

The topographic features of the VO₂ films were analyzed by atomic force microscopy (AFM) measurements, carried out with a Veeco Dimension 3100 Atomic Force Microscope.

Both continuous and nanostructured Ni films (with a thickness of 20 nm) were deposited at room temperature onto the Al/VO₂/Al₂O₃

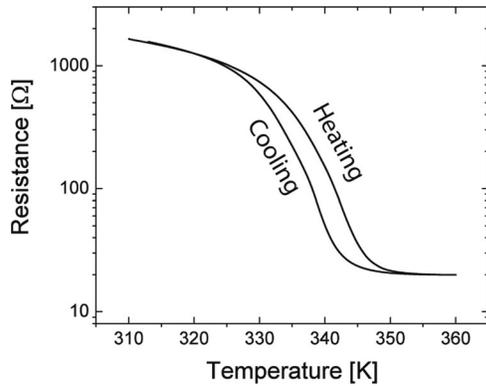


Fig. 1. Electrical resistance of the thin VO₂ films analyzed as a function of the temperature. A clear change in the resistivity of the VO₂ can be observed at the MIT temperature. For these VO₂ films, a MIT temperature of about 330 K can be identified. A clearly hysteretic behavior can be observed showing that, within a certain temperature range, bistability can be achieved.

samples by electron beam evaporation in an ultra-high vacuum chamber with a base pressure in the mid 10^{-10} mbar regime. For the fabrication of the nanostructured Ni elements, a lift-off lithography process was employed using a positive electron beam resist layer (poly(methyl-methacrylate) and co-polymer) coated on top of the VO₂ films. We used electron beam lithography (Raith PIONEER) to expose the photoresist using an electron energy of 10 keV and an exposure dose of about $120 \mu\text{C cm}^{-2}$. It was then developed for 30 s in a 1:3 solution of methyl-isobutyl-ketone and isopropanol, followed by rinsing in pure isopropanol (immersion) for 60 s. The lift-off was carried out by immersion in pure acetone.

The magnetic properties of the continuous Ni films were characterized as a function of the temperature by longitudinal magneto-optical Kerr effect (MOKE) magnetometry, employing a low-noise red laser diode (Coherent ULN-635, wavelength of 635 nm), and a variable temperature cryostat (Oxford Instruments Microstat He2) to heat the samples up to a temperature of about 400 K. For each temperature step, the temperature of the heat bath was allowed to completely stabilize before acquiring the magnetic hysteresis loops.

The magnetic configuration of the Ni nanostructured elements was characterized by quasi-static high-resolution X-ray photo-emission electron microscopy (PEEM). These measurements were carried out at the SIM (X11MA) beamline of the Swiss Light Source [Flechsig 2010, Le Guyader 2012], which is equipped with an Elmitec photoemission electron microscope (type LEEM III). The magnetic contrast in the images was achieved using X-ray magnetic circular dichroism (XMCD) tuned to the Ni L₃ edge (about 852 eV). The samples were mounted on a sample holder equipped with a resistive heater, and the temperature of the samples was controlled by a built-in thermocouple sensor. Due to the limited amount of X-ray beamtime available, only a limited number of heating/cooling cycles could be carried out. The structured Ni elements were imaged at zero applied external magnetic field.

III. RESULTS AND DISCUSSION

Fig. 1 shows that the VO₂ thin films undergo a MIT at a temperature of about 330 K, which corresponds to a two order of magnitude

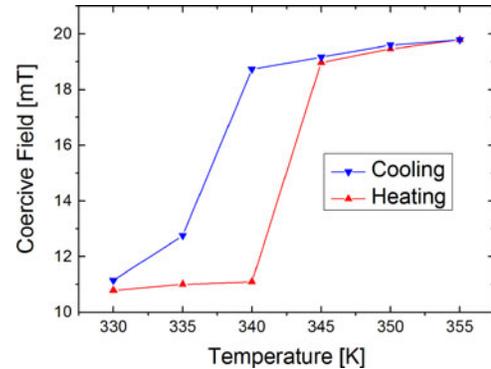


Fig. 2. Coercive field of a continuous, 10 nm thick, Ni layer grown on top of the VO₂ thin films as a function of the temperature. A clear increase of the coercive field upon crossing the MIT can be observed, along with a hysteretic behavior with respect to the temperature of the VO₂ film.

resistance change with increasing temperature. Note that the magnitude of the resistance change is reduced due to shorting by the 2 nm Al capping layer and, taking this into account, the amplitude is consistent with the expected change. The MIT threshold temperature is lower than the bulk value of about 340 K [Morin 1959]. This is due to the epitaxial straining of the thin VO₂ film caused by the growth on the substrate, as already observed for the growth of VO₂ films on different substrates [Aetukuri 2013, Muraoka 2013, Nagashima 2007, Pouget 1975].

The MIT is coincident with the SPT in the VO₂ lattice structure [Cavalleri 2001, de la Venta 2013, Viswanath 2011, Zylbersztein 1975]. This generates a change in the mechanical stress in the VO₂ film, of the order of 10^8 Pa [Viswanath 2011], which is then transferred to the thin film grown above the VO₂ layer [de la Venta 2013, Viswanath 2011], giving rise to strain variations on the order of $\epsilon \approx 10^{-3}$ for materials such as Ni (Young's Modulus $Y = 220$ GPa [Ledbetter 1973]).

As shown in Fig. 2, in the continuous film we observe an increase in the coercivity of the Ni film after heating the VO₂ above its SPT temperature, compatible with previous observations for room-temperature Ni grown on VO₂ films [de la Venta 2013]. We also observe the hysteretic behavior of the coercive field with respect to the sample temperature upon heating and cooling, which is consistent with the hysteresis of the MIT observed in the resistance measurements (see Fig. 1). Finally, we repeated the measurements for different orientations of the magnetic field, and we observed a similar increase of the coercive field for the different magnetic field directions upon crossing the SPT. The straining of the Ni film caused by the heating of the VO₂ above its SPT temperature generates magnetic anisotropy through the inverse magnetostrictive effect, which leads to a coercivity enhancement. Based on the results presented in de la Venta [2013], it is possible to estimate, for a change in the coercive field of Ni of about 5 mT, strain variations on the order of $\epsilon \approx 10^{-3}$, compatible with previous investigations [de la Venta 2013]. Note that we did not include, in the estimation of the change of the coercive field, the effect of the substrate clamping. This approximation was done taking into consideration the relatively high thickness of the VO₂ film (100 nm). While a full characterization of the influence of the VO₂ film thickness on the magnetic properties of the Ni film deposited above is beyond the scope of this letter, a larger influence of the substrate clamping—thus leading

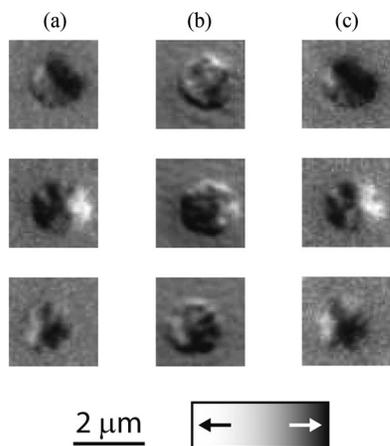


Fig. 3. XMCD-PEEM images at different temperatures of the magnetic domain configuration of three nanostructured Ni disks (2 μm diameter) grown on top of a VO_2 film. (a) Initial state at 300 K. (b) Magnetic configuration after heating the sample to about 370 K. (c) Magnetic state after cooling down again to 300 K, showing a magnetic state equal to the one presented in (a). The grayscale bar indicates the direction of the magnetic contrast in the images.

to a reduction of the transferred strain—is expected for a thinner VO_2 film.

We fabricated Ni nanostructures on top of VO_2 to probe the effect of the SPT on the domain structure of the confined magnetic states. Fig. 3(a) shows that the Ni nanostructures exhibit magnetic states that are dominated by the surface topography of the VO_2 film, due to the relatively rough growth of the Ni films on the VO_2 , which was not optimized in this work. To probe if the states can be reversibly manipulated when crossing the SPT, we used the non-trivial magnetic states observed for the Ni nanostructures. We compared the initial room temperature magnetic states [Fig. 3(a)] with those for a temperature of about 370 K [Fig. 3(b), well above the SPT], and those at room temperature after cooling from 370 K [Fig. 3(c)]. As can be seen from Fig. 3, the SPT leads to unambiguous modifications of the magnetic configurations of the structured Ni elements. As a control experiment, we carried out the same heating on a similar Ni sample (without a VO_2 film underneath) at the same temperatures, which showed only a reduction of the magnetic contrast compatible with the reduction of the saturation magnetization of the Ni at higher temperatures, compatible with what observed in previous works [de la Venta 2013]. Upon the cooling of the Ni/ VO_2 sample below the MIT threshold temperature, the original magnetic configurations of the Ni elements are recovered, as can be observed by comparing Fig. 3(a) and (c). These changes in the magnetic configuration of the Ni nanostructures are largely reproducible and reversible upon a renewed heating of the Ni/ VO_2 sample above the MIT threshold temperature.

To understand the reason for the irregularity of the observed magnetic states combined with the existence of different states in otherwise equal magnetic nanostructures, we carried out topographic AFM measurements of the as-grown VO_2 thin films, as shown in Fig. 4. We observed strong topographic features on the surface of the VO_2 films, with a surface roughness comparable to the nominal thickness of the nanostructured Ni elements. Furthermore, some of the structured Ni elements that were analyzed did not exhibit a full recovery of the original magnetic configuration upon multiple excitations of the VO_2

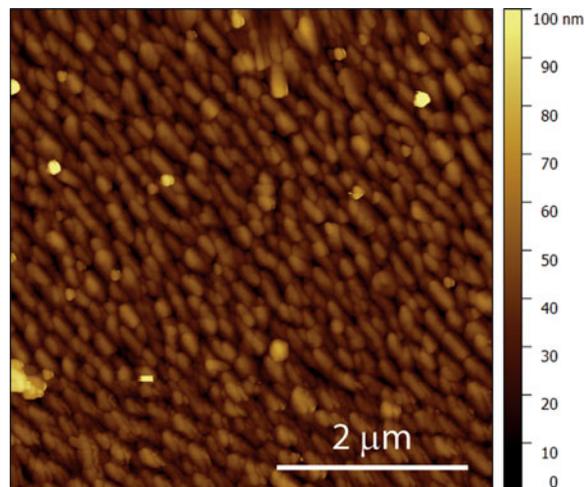


Fig. 4. Topographic AFM image of the surface of the VO_2 thin films, exhibiting a RMS surface roughness of about 15 nm, giving rise to a strong topographical influence on the magnetic configuration of the nanostructured Ni elements.

SPT. This can be attributed to the presence of magnetic pinning sites induced by the topographical features of the VO_2 film. This issue is, however, expected to be greatly reduced for structured Ni elements fabricated on top of VO_2 films with a lower surface roughness.

However, despite the influence of the local topography on the magnetic configuration of the nanostructured Ni elements, which could lead to the presence of numerous magnetic pinning sites, the changes in the magnetic state of the nanostructures induced by the crossing of the MIT are both largely reversible and reproducible. This fulfills the two necessary conditions for time-resolved measurements based on the pump-probe approach and the use of the effect for non-volatile switching. Given that we can already observe the reversible switching even for our high pinning system, with an improvement of the surface quality of the VO_2 thin films it will be possible to fabricate Ni nanostructures exhibiting shape-anisotropy dominated states, which can then be employed as starting configurations for time-resolved experiments aimed at the analysis of the dynamics of the magneto-elastic effect.

IV. CONCLUSION

We have shown that the nanostructured ferromagnetic elements on top of VO_2 thin films exhibit magnetic structures that have substantial, reversible and reproducible modifications of their magnetic configuration due to the SPT. The strong SPT dependence occurs even though the structures are highly-pinned, and it is clear that in better-defined shape-anisotropy-dominated structures the effect of VO_2 SPT will be even more pronounced. These results constitute a first key step for the analysis of the dynamical processes involved in the magneto-elastic coupling. For future work, it is still necessary to improve the surface quality of the VO_2 films so the magnetic domain configuration of the nanopatterned ferromagnets will not be affected by the topographic features of the underlying layers. Such improvements could include the fabrication of the VO_2 film at lower deposition rates, or with different techniques, such as pulsed laser deposition [Aetukuri 2013].

However, given the clear change in the spin structure, this bodes well for using ultra-fast structural phase transitions to reversibly and reproducibly switch the magnetization at the short timescales relevant for applications.

ACKNOWLEDGMENT

This is a highly collaborative research project. The experiments were conceived jointly between the Johannes Gutenberg University Mainz and the University of California San Diego, the data were extensively debated, and the manuscript was written by multiple interactions between all the co-authors. The VO₂ films were deposited and initially characterized at the University of California San Diego (IV, IKS). The fabrication of the Ni structures, along with their initial characterization, was carried out at the Johannes Gutenberg University Mainz (SF, MV, RR, RLC, MK). Part of this work was carried out at the SIM (X11MA) beamline of the Swiss Light Source, Villigen PSI, Switzerland (SF, MV, RLC, AK, MK). The authors further acknowledge the financial support from the EU 7th Framework Programme IFOX (Grant No. NMP3-LA-2010 246102), MAGWIRE (Grant No. FP7-ICT-2009-5 257707), CALIPSO (Grant No. FP7/2007-2013 312284), the European Research Council (Grants MASPIC ERC-2007-StG 208162 and MULTIREV ERC-2014-PoC 665672), the Graduate School of Excellence “Materials Science in Mainz” (Grant No. GSC 266), the Swiss National Science Foundation (SNF), the Deutsche Forschungsgemeinschaft (DFG), including the Grant SFB/TRR 173, and Department of Energy’s Office of Basic Energy Science, DMR grant DE FG02 87ER-45332 (IV, IKS), and by U.S. Air Force Office for Scientific Research grant FA9550-12-1-0381 (IV, IKS). IKS acknowledges a U.S. Department of Defense (DoD) Vannevar Bush Fellowship.

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