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1        **Strong magnetic anisotropy of epitaxial PrVO<sub>3</sub> thin films on**  
2        **SrTiO<sub>3</sub> substrates with different orientations**

3        D. Kumar,<sup>1\*</sup> P. Boullay,<sup>1</sup> A. Fouchet,<sup>1</sup> A. David,<sup>1</sup> A. Pautrat,<sup>1</sup> W. Prellier<sup>1\*</sup>

4                    <sup>1</sup>*Laboratoire CRISMAT, CNRS UMR 6508,*

5                    *ENSICAEN, Normandie Université,*

6                    *6 Bd Maréchal Juin, F-14050 Caen Cedex 4, France*

Abstract

We have probed the structural and magnetic properties of PrVO<sub>3</sub> (PVO) thin films grown on the (001)-, (110)- and (111)-oriented SrTiO<sub>3</sub> (STO) substrates. By changing the substrate orientation, (1) the out-of-plane orientation of film can be tuned to [110], [100] / [010], and [011] / [311], (2) the number of crystal variants in the film can be varied, for ex. we observe single domain film on (110)-oriented STO, whereas two domains in the film grown on the (111)-oriented STO substrate. The lattice strain induced by using different oriented substrates has direct influence on the magnetic properties of PVO films. The magnetic moment of PVO films radically enhances from 0.4  $\mu_B$  /f.u. for STO (001) to 2.3  $\mu_B$  /f.u. for STO (111). While, films on (001)-oriented STO substrate display out-of-plane anisotropy, an in-plane anisotropy is observed for films grown on the (110)- and (111)-oriented STO substrates. In addition, a strong uniaxial magnetic anisotropy is also extracted for a partially relaxed film on the (110)- oriented STO substrate. Such findings can help oxide community in the better understanding of magnetic anisotropy in vanadate thin films, a subject that still lacks scientific investigations.

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\*deepak.kumar@ensicaen.fr, wilfrid.prellier@ensicaen.fr

## 8 I. INTRODUCTION

9 The  $RVO_3$ , where  $R$  is a trivalent rare earth element ( $R = \text{La, Ce, Pr, \dots, Lu}$ ), have been  
10 intensively studied due to their wide range of physical properties [1–5]. In these compounds,  
11 the interaction between spin, orbit and lattice degrees of freedom remains the center of all  
12 intriguing physical phenomena, where one alters the ideal lattice of a compound in order to  
13 modify and even achieve new functionalities. These systems order with one of two different  
14 types of magnetic structures. For the compounds with larger rare earth radii ( $R = \text{La–Dy}$ )  
15 the magnetic structure is of C-type (where, spins order antiferromagnetically along  $ab$  plane  
16 and ferromagnetically along the  $c$ -axis), whereas compounds with smaller rare earth radii  
17 ( $R = \text{Ho–Lu}$ ) have a G-type magnetic structure (spins order antiferromagnetically in all the  
18 direction) [6].

19 Bulk  $\text{PrVO}_3$  (PVO), at room temperature, adopts an orthorhombic  $Pbnm$  crystal structure  
20 with the lattice parameters:  $a_o = 5.487 \text{ \AA}$ ,  $b_o = 5.564 \text{ \AA}$ , and  $c_o = 7.778 \text{ \AA}$  ( $o$  stands for  
21 orthorhombic) [7]. Bulk PVO is a canted antiferromagnet [8], but it has tendency to be-  
22 come a hard ferromagnet when grown under epitaxial strain in the form of a thin film [9].  
23 The impact of epitaxial strain imposed by the underlying substrate also plays a crucial role  
24 in governing the magnetism of the PVO films. Namely, when PVO films are grown on the  
25  $\text{LaAlO}_3$  (LAO) substrate, under a large compressive strain (2.9 %), the reduction in in-plane  
26 bond length between neighboring V atoms leads to promote the super-exchange interactions,  
27 and thus exhibit large Néel temperature ( $T_N$ ). While, films grown on the  $\text{SrTiO}_3$  (STO)  
28 substrate under a moderate tensile strain ( $\sim 0.1 \%$ ) show reduced  $T_N$ , in agreement with  
29 our theoretical calculations [2]. Recent advances in the technological devices allow one to  
30 realize materials in the realm of atomic resolution, and separate the effect of epitaxial strain  
31 from microstructure and defects. Through X-ray Photoelectron Spectroscopy (XPS), the  
32 formation of an over-oxidized dead-layer (4–5 nm) on the surface of PVO film was earlier  
33 revealed, consisting of isolated Pr atom. Furthermore, this dead-layer has a large param-  
34 agnetic contribution, whose impact gradually diminishes with the increase of film thickness  
35 [10]. The over-oxidation of the film surface is quite common in vanadium oxide thin films,  
36 which tends to oxidize during growth process [11, 12]. One therefore needs to be very careful  
37 during thin film-fabrication and require additional precautions to avoid the surface oxida-  
38 tion, such as using of high vacuum during growth and reduction of film surface by using a  
39 capping layer.

40 The preferential direction of magnetization in thin films is mainly determined by the mag-

41 netic anisotropy, which includes shape anisotropy, magnetoelastic anisotropy and magne-  
42 tocrystalline anisotropy. There have been numerous studies in the past to explore magnetic  
43 anisotropy in other competitive complex systems such as manganites [13–15], and ruthenates  
44 [16, 17], and due to Tunneling Magnetoresistance properties of the former, they are being  
45 used as Magnetic Tunneling Junction devices. However, little is known of the magnetic  
46 anisotropy in vanadium oxide thin films, especially PVO, and thus has yet to be under-  
47 stood.

48 In this paper, we were motivated to study the substrate surface orientation-induced mag-  
49 netic anisotropy in PVO thin films, which were grown on (001)-, (110)- and (111)-oriented  
50 STO substrates. By changing the substrate-surface orientation, the out-of-plane orienta-  
51 tion of PVO films can be effectively controlled. For the film grown on (111)-oriented STO  
52 substrate, we observe a striking enhancement in magnetization compared to its bulk coun-  
53 terpart, and, to our knowledge, is first time ever recorded. Furthermore, a strong in-plane  
54 uniaxial anisotropy is seen for film grown on the (110)-oriented STO substrate.

## 55 II. EXPERIMENTAL

56 The  $\text{PrVO}_3$  (PVO) thin films were synthesized by using Pulsed Laser Deposition (PLD) on  
57 the (001)-, (110)- and (111)-oriented  $\text{SrTiO}_3$  (STO) substrates supplied by CrysTec. A KrF  
58 excimer laser (wavelength  $\lambda = 248$  nm) with repetition rate of 2 Hz and laser fluence of 2  
59  $\text{J}/\text{cm}^2$  was focused on a  $\text{PrVO}_4$  ceramic target, with substrate-to-target distance of 5 cm.  
60 The growth temperature and pressure during deposition were maintained at 650 °C, and  
61  $10^{-6}$  mbar, respectively. X-ray diffraction and reciprocal lattice mapping were performed on  
62 a Bruker D8 Discover diffractometer (Cu  $K\alpha_1$  radiation,  $\lambda = 1.5406$  Å) to study the crys-  
63 tallinity and strain states of the epitaxial layers. The film thickness was estimated by fitting  
64 the thickness fringes around Bragg peak of substrate to the Laue equation (*Intensity of oscil-*  
65 *lations,  $I = \frac{\sin^2(NQC/2)}{\sin^2(QC/2)}$ ; where  $N$  is the number of unit cells,  $Q$  is the reciprocal lattice vector,*  
66 *and  $C$  denotes the out-of-plane lattice parameter*). The microstructure of films on (110)-  
67 and (111)-oriented STO was revealed through Transmission Electron Microscopy (TEM)  
68 (**Description of TEM instrument**). The field- and temperature-dependent magnetiza-  
69 tion measurements were performed in a Quantum Design Magnetic Property Measurement  
70 System (MPMS) XL, as well as the Vibrating Sample Magnetometer (MPMS 3).

### 71 III. STRUCTURE

72 The X-ray diffraction results of 35 nm thick PVO films grown on (001)-, (110)- and (111)-  
 73 oriented STO substrates are detailed in Fig. 1. The Laue oscillations in the  $\theta$ - $2\theta$  High  
 74 Resolution X-ray diffraction (HRXRD) patterns of PVO films on (001)- and (110)-oriented  
 75 STO are indicative of a well-defined film-substrate interfaces, whereas, hardly an oscillation  
 76 is observed for the film grown on (111)-oriented STO substrate. For STO (111) substrate,  
 77 the polar catastrophe and intermix termination could invoke major reconstructions, and this  
 78 could lead to produce rough interfaces [18]. The pseudocube interplanar spacings derived  
 79 from the film peak positions in Figs. 1(a), 1(b), 1(c), are:  $d_{001} = 3.94 \text{ \AA}$ ,  $d_{110} = 3.98/\sqrt{2} =$   
 80  $2.814 \text{ \AA}$  and  $d_{111} = 3.954/\sqrt{3} = 2.283 \text{ \AA}$ , for PVO films on (001)-, (110)- and (111)-oriented  
 81 STO substrates, respectively.

82 Reciprocal Space Maps (RSM) of PVO films on STO substrates were collected for the skew  
 83 symmetrical reflections of the substrates (Fig. 2). The inspection of these maps reveals  
 84 that the films have grown coherently on the STO substrates, with the same in-plane lattice

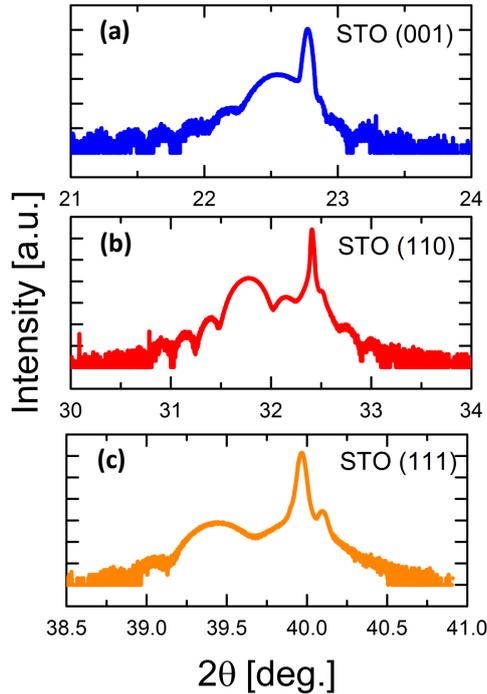


Figure 1: (Color online) HRXRD  $\theta - 2\theta$  scans of  $\text{PrVO}_3$  (PVO) thin films on (a) (001)-, (b) (110)- and (c) (111)-oriented  $\text{SrTiO}_3$  (STO) substrates, in the vicinity of  $(001)_c$ ,  $(110)_c$  and  $(111)_c$  Bragg's peak of STO, respectively. The subscript  $c$  refers to cubic.

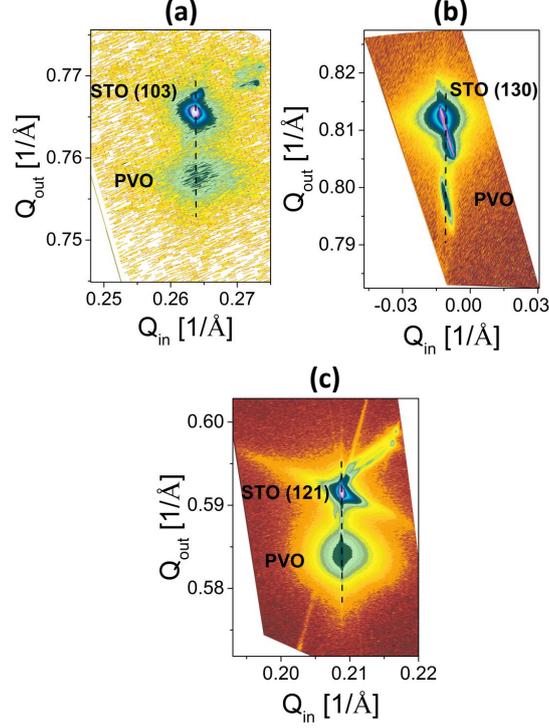


Figure 2: (Color online) X-ray diffraction reciprocal space maps around (d) STO (103), (e) STO (130) and (f) STO (121) of (001)-, (110)- and (111)-oriented STO substrates, respectively. The dashed lines are only guide to the eyes and represent strained nature of films with the substrates.

85 parameters, as the position of film peak along the horizontal  $Q_{in}$  axis matches with that of  
 86 the substrate peak for all three orientations of the substrate. Combining the HRXRD  $\theta - 2\theta$   
 87 and RSM measurements, we have calculated the pseudocube unit-cell volume of PVO:  $V_{pc}$   
 88  $= 3.905^2 \times 3.940 \text{ \AA}^3 = 60.08 \text{ \AA}^3$  for STO (001),  $V_{pc} = 3.905^2 \times 3.980 \text{ \AA}^3 = 60.66 \text{ \AA}^3$  for STO  
 89 (110) and  $V_{pc} = 3.905^2 \times 3.954 \text{ \AA}^3 = 60.29 \text{ \AA}^3$  for STO (111) ( $pc$ : pseudocubic).

90 The local microstructures of the films were investigated for films grown on (110)- and (111)-  
 91 oriented STO substrates, and the high resolution TEM results are shown in Fig. 3. For PVO  
 92 film on (111)-oriented STO substrate, two kinds of domains are observed, marked by I and  
 93 II in Fig. 3(a). The thickness of the film is estimated to be  $\sim 35$  nm, in agreement with  
 94 the XRD results. The SAED and high resolution TEM results reveal that the orientation  
 95 relations for film (F) grown on (111)-oriented STO substrate (S) are: (1)  $F[011]_o // S[111]_c$   
 96 and  $F[100]_o // S[110]_c$  (shown in red in Fig. 3(f)), or,  $F[101]_o // S[111]_c$  and  $F[010]_o //$   
 97  $S[110]_c$ , the out-of-plane lattice parameters for  $[011]_{PVO}$  and  $[101]_{PVO}$  being almost similar,  
 98 (2)  $F[311]_o // S[111]_c$  and  $F[\bar{1}\bar{1}2]_o // S[110]_c$  (shown in green in Fig. 3(g)) (the subscript  $c$   
 99 indicate cubic notation). These domains are oriented  $60^\circ$  to each other, which is consistent

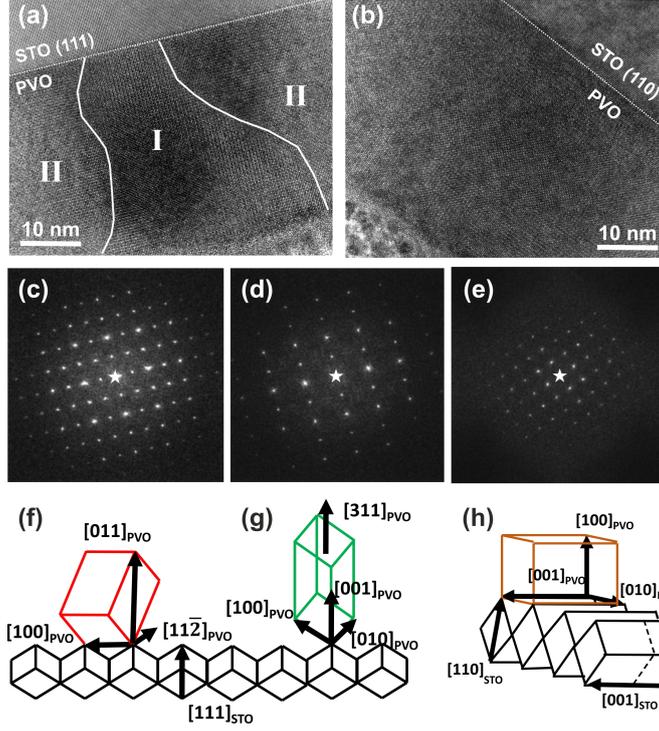


Figure 3: (Color online)(Top panel) HRTEM images of PVO films grown on (a) STO (111) and (b) STO (110) substrates. (Middle panel) (c), (d): Selected area electron diffraction (SAED) patterns for the film on (111)-oriented STO substrate, corresponding to domain I and domain II, respectively. The Zone Axis Point (ZAP) for (c) is along  $[011]_o$  and for (d) is along  $[311]_o$  (the subscript  $o$  refers to orthorhombic). (Middle panel) (e): SAED patterns for the film on (110)-oriented STO substrate, with ZAP along  $[100]_o$ . (Bottom panel) The representative schematics of the observed crystallographic domains in the film on STO (111) (f,g) and STO (110) (h).

100 with the six fold symmetry presented by the STO (111) substrate. The PVO film on (110)-  
 101 oriented STO substrate presents a sharp and parallel interface as compared to the interface  
 102 between film and STO (111) substrate (Fig. 3(b)), in agreement with the previous observa-  
 103 tions through x-ray diffraction. In addition, the TEM results confirm the thickness of film  
 104 on STO (110), to be around 35 nm. The diffraction patterns extracted from several different  
 105 regions in the film indicate presence of only one kind of domain, or, two domains but oriented  
 106  $180^\circ$  to each other. Thus, the film grows with  $[100]_o$  (or  $[010]_o$ ) as out-of-plane axis while  
 107 clamping  $[001]_o$  and  $[010]_o$  (or  $[100]_o$ ) to the substrate, with the following epitaxial relation-  
 108 ships:  $F[100]_o$  (or  $F[010]_o$ ) //  $S[110]_c$  and  $F[001]_o$  //  $S[001]_c$ . The observation of different  
 109 domains on the two substrates is correlated with the crystal symmetries of the substrates.  
 110 The orthorhombic lattice of (110)-oriented STO (see Fig. 3(h)) may allow coherent growth

111 of orthorhombic cell of PVO without any atomic reconstruction at the interface, therefore  
 112 presenting sharp interface in combination with well-defined thickness fringes in the XRD  
 113 scan. On the contrary, STO(111) substrate is known to undergo atomic reconstruction at  
 114 the interface to suppress polar discontinuity and its divergent surface energy [18, 19], making  
 115 this substrate less compatible to grow orthorhombic perovskite lattice. For PVO film grown  
 116 on (001)-oriented STO substrate, we have already observed two domains, oriented  $90^\circ$  to  
 117 each other with the epitaxial relationship:  $F[110]_o // S[001]_c$  and  $F[001]_o // S[100]_c / S[010]_c$   
 118 [2, 9]. In general, the sign and strength of lattice mismatch vary with the orientations of the  
 119 substrate, and thus different distortions may be induced in the film, yielding different crystal  
 120 structures. The calculated in-plane lattice mismatches and observed growth orientations of  
 121 the films for different oriented STO substrates are listed in Table I. The (001)-oriented STO  
 122 exerts tensile stress on PVO  $[001]_o$ -axis (see Table I), imposing tetragonal distortion on the  
 123 unit cell [2]. On the other hand, (110)- and (111)-oriented STO substrates would impose

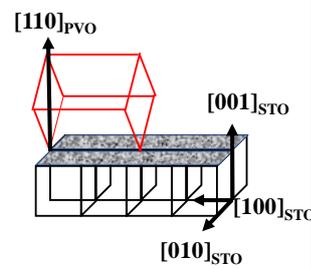
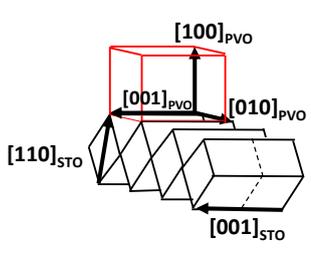
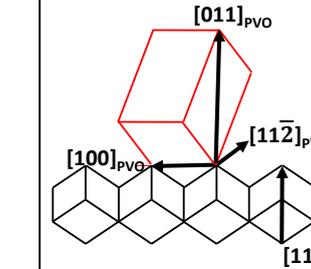
	STO[001]			STO [110]			STO [111]		
	In-plane		Out-of-plane	In-plane		Out-of-plane	In-plane		Out-of-plane
Directions	$[001]_o = [100]_{pc}$	$[\bar{1}\bar{1}0]_o = [010]_{pc}$	$[110]_o = [001]_{pc}$	$[001]_o = [001]_{pc}$	$[010]_o/[100]_o = [110]_{pc}$	$[100]_o/[010]_o = [110]_{pc}$	$[11\bar{2}]_o = [110]_{pc}$	$[100]_o/[010]_o = [110]_{pc}$	$[\bar{1}01]_o/[0\bar{1}1]_o = [111]_{pc}$
Lattice mismatch	0.41 %	-0.05 %		0.41 %	-0.74 % / 0.65 %		0.18 %	0.65 % / -0.74 %	
Growth									

Table I: (Color online) A detailed summary of the in-plane and out-of-plane lattice directions of PVO grown on STO substrates with different orientations. The lattice misfits between in-plane directions of PVO and the substrate are shown. The experimentally observed growth orientations of the PVO lattice on different oriented STO substrates are also shown. Here, a positive lattice mismatch between PVO and substrate indicates tensile stress, and negative indicates compressive stress.

124 monoclinic and trigonal distortions, respectively, due to unequal lattice mismatches along  
125 two in-plane orthogonal directions.

#### 126 IV. MAGNETISM

127 Since the strain and lattice distortion depend on the orientations of the STO substrates,  
128 the magnetic properties of PVO films are expected to differ for the three orientations of the  
129 substrate, due to change in the V–O bond length and V–O–V bond angle. The primary  
130 magnetic results of 35 nm thick PVO thin films are shown in Fig. 4. For PVO film on  
131 the (001)-oriented STO, the saturated magnetization ( $M_s$ ) is  $\sim 0.3 \mu_B / \text{f.u.}$ , and is close  
132 to our earlier observations [2]. Remarkably, the saturated magnetization for PVO films  
133 on (110)- and (111)-oriented STO substrates has extensively enhanced, close to  $1.0 \mu_B /$   
134  $\text{f.u.}$  and  $2.3 \mu_B / \text{f.u.}$ , respectively. These obtained values of  $M_s$  are larger as compared to  
135 that of the bulk, which is  $\sim 0.6 \mu_B / \text{f.u.}$  [8]. Due to the polar nature of the (110)- and  
136 (111)-oriented STO substrates, they are more susceptible to the reconstruction. This could  
137 allow a quicker recovery of the spin moments, leading to an enhanced magnetization. To  
138 the best of our knowledge, this striking phenomenon where the saturated magnetization is  
139 increased by approximately 8-fold merely by changing the crystal surface orientation of the  
140 substrate, is perhaps observed first time in the oxide thin films. Indeed, a similar study  
141 focussed on SrRuO<sub>3</sub> thin films grown on different oriented STO substrates reported nearly  
142 2 times enhancement of saturated magnetization (between (001) and (111)-oriented STO),  
143 and the authors explained this peculiar behavior using a high spin state of Ru<sup>4+</sup> ions [20].  
144 However, V<sup>3+</sup> ion has only two electrons in the 3d shell ( $t_{2g}$  orbital-active system), making  
145 them ineligible for high spin state context which typically require at least four electrons  
146 in the 3d shell. Nevertheless, a dead-layer present at the surface of film and consisting of  
147 isolated paramagnetic Pr ions, could also explain the observed substantial magnetization.  
148 In fact, we have recently shown that a dead layer of  $\sim 5$  nm in thickness, which is composed  
149 of mainly V<sup>4+</sup> ions, is responsible for the increase of magnetization for low thicknesses films  
150 [10].

151 Remarkably, the steps in the hysteresis loops (especially for STO (111)) are also seen,  
152 and are almost similar to what were previously reported for PrVO<sub>3</sub> single crystals (below  
153 3 K) [21]. In the stated reference [21], the steps (in hysteresis loop) strongly depended  
154 on the field sweep rate, namely, they shifted to a higher magnetic field when sweep rate  
155 was decreased. This unusual behavior was then proposed to be arisen on the account of

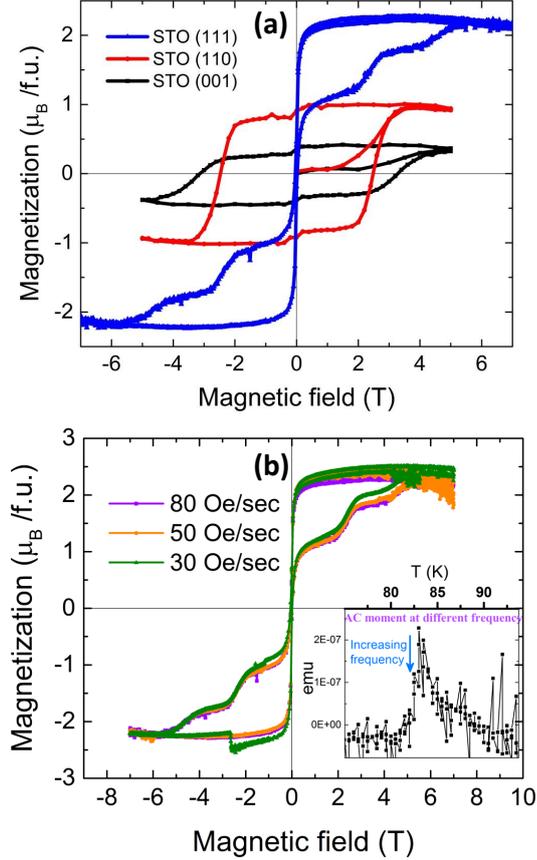


Figure 4: (a) Hysteresis cycles at 10 K for 35 nm thick PVO films on (001)-, (110)- and (111)-oriented STO substrates, obtained by applying magnetic field in the plane of sample. (b) Hysteresis cycles for PVO film on (111)-oriented substrate at different field sweep rates. Inset shows the AC moment of the same sample as a function of temperature (in the vicinity of magnetic ordering temperature) taken at different frequencies.

156 two phases in  $\text{PrVO}_3$  single crystals, one with the fraction of spins responsible for the  
 157 glassy-like behavior, and other spins ordered in the antiferromagnetic fashion. Thus, in  
 158 order to verify if the steps in the hysteresis loops are related to spin-glass-like mechanism,  
 159 we have collected hysteresis loops for PVO film on (111)-oriented STO at different field  
 160 sweep rates (Fig. 4(b)), in combination with the AC moment of the sample at different  
 161 frequencies in the vicinity of magnetic ordering temperature (inset of Fig. 4(b)). We infer  
 162 that, neither the steps in hysteresis loop depend upon field sweep rate, nor the magnetic  
 163 ordering temperature show variation as a function of frequency, a necessity for spin glass  
 164 system. These observations suggest that the steps in the hysteresis loop may not essentially  
 165 be related to the spin-glass structure, but perhaps to the magnetic domains in the film.

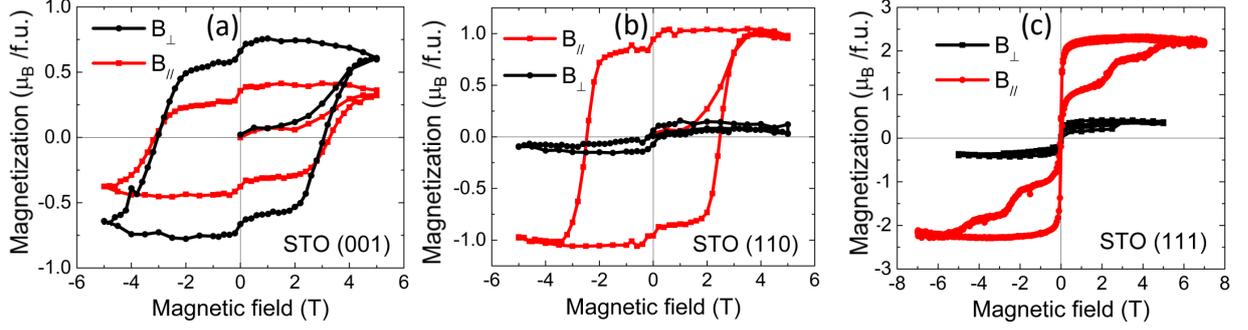


Figure 5: (a)-(c) Hysteresis cycles for films on three oriented STO substrates with field applied along the sample plane and perpendicular to the sample.

166 The magnetic anisotropy of the PVO films was characterized by  $M$ - $H$  curves after the  
 167 subtraction of the diamagnetic contribution from the oriented STO substrates. The  
 168 magnetic field was applied along in-plane and out-of-plane of the sample, and the results are  
 169 shown in Fig. 5. On one hand, for PVO film on (001)-oriented STO, the largest saturated  
 170 magnetization within the measured magnetic field range is acquired when magnetic field is  
 171 applied along out-of-plane direction, and thus regarded as the magnetic easy axis. On the  
 172 other hand, for films on (110)- and (111)-oriented STO, a strong anisotropy is observed,  
 173 where, the largest saturated magnetization is achieved for field applied along in-plane of  
 174 the sample. Therefore, the magnetic easy axis most probably remains normal to the film  
 175 plane, along PVO  $[110]_o$  for (001)-oriented STO, while it changes along the film plane for  
 176 (110)- and (111)-oriented STO, along PVO $[001]_o$  and PVO $[11\bar{2}]$ , respectively. This could  
 177 be due to the deformed  $\text{VO}_6$  octahedra, which is extended (compressed) along out-of-plane  
 178 direction for a compressive (tensile) strain. A summary of the magnetic parameters;  
 179 coercive field ( $H_c$ ), remanent magnetization ( $M_R$ ) and saturated magnetization ( $M_s$ ), for  
 180 field parallel (in-plane) and perpendicular (out-of-plane) configuration, are listed in Table II.

181

182

183 We have further investigated the effect of strain relaxation in PVO films on the uniaxial  
 184 / biaxial magnetic anisotropy. For this, films of thickness  $\sim 75$  nm were fabricated on  
 185 the (001)-, (110)- and (111)-oriented STO substrates. The x-ray reciprocal space maps  
 186 (not shown) recorded for skew symmetrical planes of the substrates show that the films are  
 187 partially relaxed. Fig. 6 shows the anisotropy results of 75 nm thick films for the three

Table II: Crystallographic orientation and angular dependence of coercive field  $H_c$ , remanent magnetization  $M_R$ , and the saturation magnetization  $M_s$  for 35 nm thick PVO films. Here,  $\theta$  (angle between direction of magnetic field and sample surface) =  $0^\circ$  means field is applied *in-plane*, and  $\theta = 90^\circ$  means field is orthogonal to sample surface.

Orientation		$0^\circ$ / <i>in-plane</i>	$90^\circ$ / <i>out-of-plane</i>
001	$H_c$ (T)	3.1	3.0
	$M_R$ ( $\mu_B$ / f.u.)	0.35	0.66
	$M_s$ ( $\mu_B$ / f.u.)	0.42	0.75
	axis	Hard	Easy
110	$H_c$ (T)	2.4	0.2
	$M_R$ ( $\mu_B$ / f.u.)	0.9	0.06
	$M_s$ ( $\mu_B$ / f.u.)	1.0	0.15
	axis	Easy	Hard
111	$H_c$ (T)	0.03	0.03
	$M_R$ ( $\mu_B$ / f.u.)	0.47	0.03
	$M_s$ ( $\mu_B$ / f.u.)	2.3	0.4
	axis	Easy	Hard

188 orientations of the substrates. For film on the (001)-oriented STO, the saturated magne-  
189 tization is largest along the out-of-plane direction of substrate (or film) (Fig. 6(a)), and  
190 could be the magnetic easy axis. For two orthogonal in-plane directions of the substrate, we  
191 observe similar shapes of the hysteresis loops, along with analogous magnetizations (both  
192 saturated and remanence). This indicates isotropic magnetism probably due to dominance  
193 of four-fold crystalline anisotropy in the PVO film on (001)-oriented STO substrate.

194 For (110)-oriented STO substrate, we observe an obvious square-like hysteresis loop with  
195 the largest magnetization along in-plane [001] of STO, and perhaps is the magnetic easy axis.  
196 While the hard axis lies along out-of-plane [110] of the STO, as evidenced by the linear hys-  
197 teresis loop (inset of Fig. 5(b)). Therefore, PVO film on the (110)-oriented STO substrate  
198 noticeably shows a perpendicular anisotropy. Interestingly, the magnetization along the two  
199 in-plane orthogonal directions differ extensively. This clearly shows the presence of uniaxial  
200 anisotropy for film on (110)-oriented STO substrate, and can be account for the magne-  
201 toelastic anisotropy present in the film due to unequal lattice mismatch along two in-plane  
202 directions. Finally, for the PVO film on (111)-oriented STO, the magnetic easy axis is along

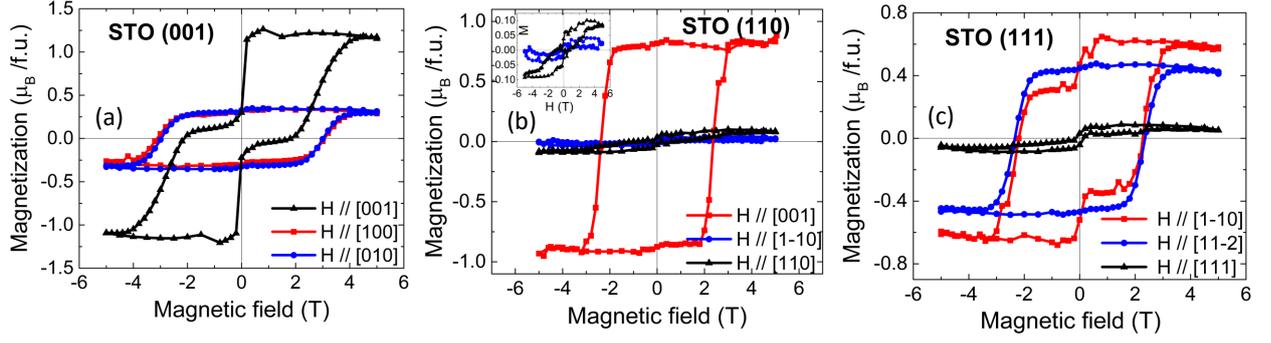


Figure 6: The hysteresis loops of 75 nm thick PVO films on (a) (001)-, (b) (110)- and (c) (111)-oriented STO substrates, along two in-plane and out-of-plane directions of the substrates.

203 any of the in-plane directions, and the hard axis remains along the out-of-plane [111]-axis  
 204 direction of the substrate. The remanences of the two in-plane directions are nearly equal,  
 205 although the saturated magnetization remains higher along  $[1\bar{1}0]$  direction. Also, given the  
 206 six-fold symmetry of the STO (111), the easy axis could lie along any of the in-plane axis  
 207 besides  $[1\bar{1}0]$  and  $[11\bar{2}]$  which stabilizes the energy balance between magnetocrystalline en-  
 208 ergy and both magnetostatic and magnetostriction energy. The deformation of the oxygen  
 209 octahedra surrounding V, elongated and compressed along the out-of-plane direction for  
 210 compressive and tensile strain, respectively, can be at the origin of the observed magnetic  
 211 anisotropy. Moreover, in the present case, the in-plane strains are unequal along the two  
 212 in-plane orthogonal directions, producing further in-plane deformation of the octahedra, and  
 213 thus causing in-plane uniaxial anisotropy. However, the direction of the magnetic easy axis  
 214 along out-of-plane (probably) for STO (001), and along in-plane for STO (110) and STO  
 215 (111), is still confusing. In general, the compressive strain enhances the out-of-plane magne-  
 216 tization, while tensile strain increases the in-plane magnetization [14, 22, 23]. Accordingly,  
 217 we have estimated an approximate pseudocube expansion (compare with bulk) of the unit  
 218 cell of PVO for the three orientations of the substrate, to be  $\sim 1.2\%$  (STO (001)),  $\sim 2.5\%$   
 219 (STO (110)) and  $\sim 1.7\%$  (STO (111)). From our earlier report [2], we know that the tensile  
 220 strain in PVO expands the unit cell, whereas, the compressive strain effectively reduces the  
 221 unit cell volume of PVO. Therefore, in this sense, STO (110) and STO (111) substrates  
 222 probably impose more tensile strain on the unit cell of PVO, forcing the magnetic easy axis  
 223 to stay along in-plane direction. On the other hand, film on STO (001) can favor a weak  
 224 out-of-plane anisotropy (present case), or no anisotropy at all (in Ref. [1]) due to a negligible  
 225 tensile strain .

## 226 V. CONCLUSIONS

227 In summary, we have studied the magnetic anisotropy of PrVO<sub>3</sub> thin films on the oriented  
228 SrTiO<sub>3</sub> substrates. We have found that the magnetization of PrVO<sub>3</sub> thin film on (111)-  
229 oriented SrTiO<sub>3</sub> substrate can be extensively enhanced compare to the bulk. All the films  
230 clearly show magnetic anisotropy. While films on the (001)-oriented SrTiO<sub>3</sub> show a weak  
231 out-of-plane anisotropy, a strong in-plane anisotropy is observed for films grown on the  
232 (110)- and (111)-oriented SrTiO<sub>3</sub> substrates. Furthermore, an evident uniaxial anisotropy  
233 is also argued for films on the (110)-oriented SrTiO<sub>3</sub> substrates, and would probably require  
234 further experiments, e.g., XPS/XAS, X-ray Magnetic Circular Dichroism (XMCD) in order  
235 to confirm a dead layer on the surface of the films (especially for film on STO(111)) and to  
236 observe the magnetism from individual cations, respectively.

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