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Magnetic domain wall contrast under zero domain contrast conditions in spin polarized low energy electron microscopy

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Authors

Zhou, Chao

Chen, Gong

Xu, Jia

et al.

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32 1. Introduction

33 Spin polarized low energy electron microscopy (SPLEEM) is a special kind of low
34 energy electron microscopy (LEEM) that uses a spin polarized electron beam to
35 generate magnetic domain images from samples [1,2,3]. Due to its high spatial
36 resolution, SPLEEM provides a valuable means to study magnetic domain structures in
37 surfaces [4] and thin films [5,6,7,8], especially in systems near the spin reorientation
38 transition [9,10,11,12], as well as to study magnetization profiles in magnetic
39 nanoparticles [13,14,15]. The capability to image the orientation of the magnetization
40 vector with high angular resolution makes SPLEEM a particularly useful tool for
41 studying domain wall (DW) spin textures [16,17,18,19]. The energy dependence of
42 electron reflectivity at surfaces is a consequence of the unoccupied electronic structure
43 of the sample, and applying SPLEEM to measure electron reflectivity spectra can be
44 used to determine spin-dependent electronic structures [2,20], as was demonstrated
45 using quantum well states in thin films [21,22,23,24,25].

46 High lateral resolution in SPLEEM makes it possible to study energy spectra in
47 non-collinear spin textures such as in magnetic DWs. Due to spin-orbit coupling, the
48 electronic structure of non-collinear spin textures can be different from that of collinear
49 ones. Recently, C. Hanneken et al. demonstrated that non-collinear spin textures in
50 magnetic skyrmions can alter the local electronic structure, resulting in a tunneling
51 resistance different from magnetic single domains [26]. More generally, the extent to
52 which the local electronic structure inside magnetic DW may be modulated by the non-
53 collinear spin texture remains an interesting question, and SPLEEM measurements may
54 provide experimental evidence.

55 Aiming to investigate the influence of non-collinear spin structures on local
56 electronic structure, this study was initiated by measuring energy dependent spectra in
57 several magnetic thin films. In most magnetic film systems, the spin asymmetry signal
58 of the electron reflectivity spectra varies as a function of energy. We found an unusual
59 contrast in magnetic DWs at the electron landing energies where domain contrast from
60 the spin dependence of the electron reflectivity vanishes. This effect is found to be
61 independent of the magnetic materials and substrates. This DW contrast is also

62 independent of the spin rotation chirality inside the DW, but is related to the slope of
63 spin asymmetry spectrum at this energy. Our analysis shows that this effect is caused
64 by the combination of finite energy spread of the electron beam and the dispersion of
65 the magnetic prism array used in the instrument to separate illumination- and imaging
66 columns. Our simulation results are in good agreement with the experimental
67 observations. The discovery of such an instrumental artefact is important for the
68 analysis of magnetic domain structure and is useful to aid correct interpretation of
69 SPLEEM images.

70

71 **2. Experiments**

72 Our experiments were performed in the SPLEEM systems at the National Center
73 for Electron Microscopy at the Lawrence Berkeley National Laboratory [1]. The
74 SPLEEM measurements were performed on Co/W(110), (Co/Ni)_n/W(110) and
75 Co/Cu(001) thin film/multilayer systems. The W(110) substrate was cleaned by cycles
76 of flash heating to 1950 °C in 3×10⁻⁸ Torr O₂ background followed by a final annealing
77 at the same temperature under ultrahigh vacuum [14]. The Cu(001) substrate was
78 cleaned by several cycles of Ar-ion sputtering and annealing to 550 °C, followed by a
79 final annealing at the same temperature in ultrahigh vacuum [12]. Co and Ni films were
80 deposited at room temperature from electron beam evaporation sources. The film
81 thickness was controlled by monitoring the electron reflectivity oscillations associated
82 with atomic layer-by-layer growth.

83 All the SPLEEM images were measured at room temperature on the as-grown
84 samples which were also prepared at room temperature. During the SPLEEM
85 measurements, the sample was biased with a start voltage (V_s), which determines the
86 electron landing energy [2]. The magnetic contrast, which strongly depends on the start
87 voltage V_s , is determined by measuring the spin-dependent reflectivity asymmetry A
88 between the spin-polarized beams with opposite polarization. SPLEEM images are then
89 calculated pixel-by-pixel using $A=[I_{\uparrow}(V_s)-I_{\downarrow}(V_s)]/[I_{\uparrow}(V_s)+I_{\downarrow}(V_s)]$ where $I_{\uparrow}(V_s)$ and $I_{\downarrow}(V_s)$
90 are the electron reflectivities with up and down spin polarization. To enhance image
91 quality, sequences of images are acquired under identical conditions and residual

92 thermal drift is corrected using image cross-correlation software. Averaging such drift-
93 corrected image sequences results in signal-to-noise optimized images. For a particular
94 sample, the magnetic contrast A in a SPLEEM image is known to be proportional to the
95 product of local unit-magnetization vector \mathbf{m} and electron beam polarization σ , i.e.
96 $A(V_s)=c(V_s)\mathbf{m}\cdot\sigma$, where the coefficient $c(V_s)$ strongly depends on the start voltage V_s .
97 The magnetic contrast A is zero for nonmagnetic samples ($\mathbf{m}=0$) and for magnetic
98 samples with \mathbf{m} perpendicular to σ ($\mathbf{m}\perp\sigma$). On the other hand, the coefficient $c(V_s)$ is
99 related to the electronic band structure of the sample, and could cross zero at certain V_s ,
100 in such cases the magnetic contrast A can be zero even for the magnetic sample with σ
101 parallel to \mathbf{m} ($\mathbf{m}\parallel\sigma$) [18-22]. During the SPLEEM measurements, we first precisely
102 adjusted the spin orientation σ to be aligned parallel to the magnetization orientation \mathbf{m}
103 in imaged domains, then we measured the energy-dependent asymmetry spectra $A(V_s)$
104 of the same magnetic domains. From the measured $A(V_s)$ spectra, we determined values
105 of V_{s0} with zero domain contrast $A(V_{s0})$. If the electronic structure indeed is modulated
106 by non-collinear spin structures, then it is expected that the $A(V_s)$ spectra inside the DW
107 would be different from that in the collinearly magnetized domains. Specifically, one
108 would expect that the zero-contrast conditions $A(V_{s0})$ might be shifted in energy, so that
109 at start voltage V_{s0} with zero contrast in the domains it is possible to observe the
110 magnetic contrast inside the DW. In search of such band structure effects we carefully
111 investigated the magnetic contrast $A(V_s)$ in DWs – compared to domains, as a function
112 of V_s .

113 3. Experimental results

114 We first present the typical measurements from a 4 monolayer (ML) Co film grown
115 on W(110) substrate, as shown in Figure 1. Co/W(110) films have strong in-plane
116 uniaxial magnetic anisotropy [27], consequently there are only two types of magnetic
117 domains separated by 180-degree DWs. The LEEM image in the inset of Fig. 1(a)
118 shows clear atomic steps, which demonstrates high quality of this film. Moreover,
119 during the Co film growth, the electron reflectivity shows clear intensity oscillation
120 [24], which supports the layer-by-layer growth of Co film.

121 The measured reflectivity spectrum $R(V_s)$ and spin asymmetry spectrum $A(V_s)$ with

122 $\mathbf{m} // \sigma$ both exhibit clear oscillations due to the electron Fabry-Pérot interference in the
 123 Co film [18-22], as shown in Figs. 1(a) and 1(b). The asymmetry spectrum $A(V_s)$ also
 124 shows clear sign reversal, which causes the domain contrast to alternate as a function
 125 of V_s . For example, the SPLEEM domain images acquired with $V_s=4.5$ V and 6.2 V
 126 have opposite contrast, although the domain contrast for $V_s=4.5$ V is smaller than that
 127 for $V_s=6.2$ V, as shown in Fig. 1(c) and 1(d). The $A(V_s)$ spectrum in Fig. 1(b) shows
 128 zero asymmetry signal at $V_{s0}=4.9$ V and 6.9 V, which indicates that at these electron
 129 energies no contrast should be visible between the domains even when the spin
 130 polarization is parallel to the magnetization. Moreover, magnetic contrast within DWs
 131 should vanish under these imaging conditions, both because of the vanishing
 132 asymmetry signal and, in addition, because orthogonal alignment between the
 133 magnetization \mathbf{m} and the beam polarization σ within the DWs cause the asymmetry
 134 $A(V_s)=c(V_s)\mathbf{m} \cdot \sigma$ to vanish at all energies. However, SPLEEM measurements at $V_s=4.9$
 135 V and 6.9 V show a surprising result: we find that unusual bright or dark contrast
 136 appears along DWs, as shown in Figs. 1(e) and 1(f). For $V_s=4.9$ V, the left DW in Fig.
 137 1(e) has dark contrast, and the right DW has bright contrast. For $V_s=6.9$ V, this contrast
 138 in the two DWs is reversed. The strength of this DW contrast at these two energies can
 139 be quantified, as shown in the line profiles plotted in Fig. 1(g).

140 In SPLEEM experiments the spin orientation of the incident electron beam can be
 141 adjusted to point along any direction (this enables the technique's vector imaging
 142 capability to resolve DW spin textures [16,17,18,19]). We adjusted the incident electron
 143 spin perpendicular to the magnetization in the domain, i.e. $\mathbf{m} \perp \sigma$: imaging with this spin
 144 alignment confirms that the DW spin texture in the Co/W(110) system is Néel-type [28]
 145 with the magnetization inside the DWs lying in the film plane. Figure 2(a) and 2(b)
 146 show such SPLEEM images, measured at $V_s=4.5$ V and 6.2 V, respectively. These two
 147 images show DWs with opposite magnetic contrasts as a result of the sign reversal of
 148 the asymmetry $A(V_s)$ between these energies, see Fig. 1(b). The 180° DWs appear in
 149 black or white contrast, depending on the rotation sense of the in-plane spin texture
 150 within the DW. However, the new DW contrast that we observe at electron energies
 151 $V_s=4.9$ V and 6.9 V, as shown in Fig. 1(e) and 1(f), appears in a homogeneous black or

152 white contrast, independent of the rotation sense of the DW spin texture. Moreover, we
 153 performed SPLEEM measurements at $V_s=4.9$ V and 6.9 V with $\mathbf{m}\perp\boldsymbol{\sigma}$. Although $c(V_s)$
 154 is zero at $V_s=4.9$ V and 6.9 V, a new kind of DW contrast is now observed within the
 155 DWs [Figs. 2(c) and 2(d)]. Different from the uniform contrast in the case of $\boldsymbol{\sigma}\parallel\mathbf{m}$, as
 156 shown in Fig. 1, in the case of $\mathbf{m}\perp\boldsymbol{\sigma}$ alternating black and white contrast now traces the
 157 DWs. This DW contrast also reverses sign for $V_s=4.9$ V versus 6.9 V respectively. The
 158 alternating contrast across the DWs can be quantified by line profiles as shown in Fig.
 159 2(e). It should be noted that the DW contrast, as shown in Fig. 2(c) and 2(d), also
 160 reverses sign as a function of the rotation sense of the in-plane spin texture within the
 161 DW.

162 To understand this unusual DW contrast, we focus on the observation that in the
 163 condition $\mathbf{m}\parallel\boldsymbol{\sigma}$ this contrast does not depend on the rotation sense of the magnetization
 164 within the DW, see SPLEEM images in Figs. 1(e) and 1(f). This suggests that the
 165 contrast may be related to the DW magnetization component orthogonal to the electron
 166 spin, or it may be related to the magnetization gradient across the DW. In order to
 167 investigate the relation between the observed DW contrast and the magnetization
 168 component orthogonal to the electron beam spin polarization, we performed the
 169 experiments on the Co(15 ML)/Cu(001) system. This system has an in-plane four-fold
 170 magnetic anisotropy [29], thus it contains 90° DWs. Fig. 3(a) and 3(b) show the
 171 reflectivity spectrum $R(V_s)$ and the spin asymmetry spectrum $A(V_s)$ respectively. A
 172 sign reversal of the asymmetry occurs at $V_{s0}\sim 7.8$ V. Although the Co/Cu(001) system
 173 has four-fold symmetry, an additional two-fold symmetry is due to the small miscut of
 174 the substrate, which distinguishes the step array in the LEEM image shown in Fig. 3(a)
 175 inset. Due to this step-induced two-fold symmetry, 180° DWs dominate the magnetic
 176 domain structure in the as-grown sample [30]. SPLEEM image at $V_s\sim 10.5$ V with the
 177 spin polarization aligned along Cu[110] (Fig. 3d), clearly shows grey areas between the
 178 white and dark regions. When the spin polarization is rotated by 90° , i.e. spin
 179 polarization aligned along Cu[$\bar{1}\bar{1}0$] (Fig. 3e), the areas that are grey in Fig. 3(d) now
 180 show strong white and black contrast in Fig. 3(e). Taken together, the SPLEEM images
 181 in Figs. 3(d,e) establish magnetization directions as indicated by the arrows in Fig. 3(d),

182 i.e. the smaller domain near the middle of the images is magnetized along Cu[1 $\bar{1}$ 0], and
183 it is separated by 90° DWs from the large domains magnetized along Cu[110]. As in
184 the case of Co/W(110) described above, we find again the similar DW contrast in
185 SPLEEM images of Co/Cu(100) acquired at electron energy with zero spin contrast,
186 i.e. at $V_s \sim 7.8$ V. For electron beam spin polarization aligned along Cu[110], we observe
187 the DW contrast at the 180° DWs, as indicated by the line profiles A and B in Fig. 3(c).
188 A similar contrast can also be observed at 90° DWs, see Fig. 3(f), although it is weaker
189 than that at 180° DWs; the spin contrast at 90° DWs is more clearly confirmed by the
190 line profile C shown in Fig. 3(c). No additional contrast is observed within the small
191 domain where the magnetization is perpendicular to the spin polarization in Fig. 3(f).
192 This indicates that the usual DW contrast in Fig. 1 and Fig. 3 is not related to the
193 orthogonal relation between the spin polarization σ and the magnetization m within
194 DWs.

195 We now consider the possibility that the unusual DW contrast may be related to the
196 magnetization gradient across the DWs. An unconventional gradient effect in magnetic
197 DW imaging has also been reported in magneto-optic microscopy studies [31,32]. R.
198 Schafer and A. Hubert first reported this new magneto-optic effect related to non-
199 uniform magnetization on the surface of a ferromagnet, and found an unexpected
200 alternating domain boundary contrast not related to the internal structure of the DWs
201 [33]. This magneto-optic gradient effect has been explained by diffraction theory
202 [34,35]. One may conjecture that the unconventional SPLEEM DW contrast is
203 proportional to the magnetization gradient, i.e. $\propto \nabla(\mathbf{m} \cdot \sigma)$. This would be consistent
204 with the fact that the unconventional DW contrast is independent of the magnetization
205 rotation sense inside DWs in Fig. 1 and Fig. 3, and it is consistent with the observed
206 opposite contrast in the neighboring DWs, which have opposite magnetization gradient.
207 Magnetization gradient dependent contrast can also explain the alternating black and
208 white DW contrast in images acquired with the spin polarization perpendicular to the
209 magnetization in the wall.

210 The proposed link with the DW magnetization gradient suggests that investigating
211 the dependence of the unconventional DW contrast on varying DW orientation may

212 yield key clues. So, we further studied the unconventional DW contrast in $(\text{Co/Ni})_n$
213 multilayers grown on W(110). This system contains many small domains with domain
214 sizes comparable to the SPLEEM field of view ($10\ \mu\text{m}$ in this work), which allows us
215 to study the dependence of the DW contrast on continuously varying DW orientation
216 [8,13]. Moreover, in the $(\text{Co/Ni})_n/\text{W}(110)$ system, the in-plane easy axis can be tuned
217 from $[001]$ to $[1\bar{1}0]$ by adjusting the relative thickness ratio of Co and Ni layers [8].

218 Figure 4 shows SPLEEM images of $\text{Co}(2)/[\text{Ni}(2)/\text{Co}(1)]_2/\text{W}(110)$ and
219 $[\text{Co}(1)/\text{Ni}(3)]_2/\text{W}(110)$ with the easy axis along $[001]$ and $[1\bar{1}0]$ respectively
220 (numbers inside round brackets indicate the corresponding film thickness in units of
221 atomic monolayer). Figs. 4(a) and 4(b) show the corresponding asymmetry spectra with
222 multiple sign reversals. Fig. 4(c) shows a SPLEEM image of
223 $\text{Co}(2)/[\text{Ni}(2)/\text{Co}(1)]_2/\text{W}(110)$, acquired at $V_{s0}=5.25\ \text{V}$, where $A(V_{s0})=0$ and thus no
224 domain contrast is observed. Similarly, Fig. 4(d) shows a SPLEEM image of
225 $[\text{Co}(1)/\text{Ni}(3)]_2/\text{W}(110)$ with zero domain contrast at $V_{s0}=5.35\ \text{V}$. These images, panels
226 4(c) and 4(d), provide a clue to the origin of the observed DW contrast: note that in Fig.
227 4(c) the DW contrast is white in the left DW and black in the right DW, while in Fig.
228 4(d) the DW contrast is reversed. Noting the opposite slopes of the corresponding $A(V_s)$
229 curves in Fig. 4(a) and 4(b), as indicated by the red arrows, this suggests a possible
230 relationship between the observed new DW contrast and the slope of the asymmetry
231 curves $A(V_s)$. An additional clue is the observation of vanishing DW contrast within
232 DW sections aligned approximately horizontally in Fig. 4(c) and 4(d), as highlighted
233 by the red oval surrounding the bottom horizontal DW sections. On the top horizontal
234 DW sections, the DW contrast gradually changes its sign, but the contrast vanishing
235 behaviour is less obvious, since the DW orientation there changes more rapidly. We
236 analysed DW contrast visibility quantitatively, in terms of asymmetry A , as a function
237 of the wall orientation angle θ_{DW} . The $A(\theta_{\text{DW}})$ curves of both samples, reproduced in
238 Figs. 4(e) and 4(f), can be well described by the relation $A_0\sin(\theta_{\text{DW}}-\theta_0)$, where A_0 is the
239 maximal value of this DW contrast and θ_0 as the offset angle with the zero contrast.
240 The best-fit value of θ_0 in both measurements is 7° and 27° respectively. Although the
241 in-plane easy magnetization axis in $\text{Co}(2)/[\text{Ni}(2)/\text{Co}(1)]_2/\text{W}(110)$ is rotated by 90°

242 compared to $[\text{Co}(1)/\text{Ni}(3)]_2/\text{W}(110)$, in both samples zero contrast of the DWs occurs
243 when the DWs are nearly parallel to the x-axis. This indicates that the observed DW
244 contrast is independent of both the magnetization orientation and the crystal axis
245 orientation. If this DW contrast were to originate from the intrinsic magnetic properties
246 of the system, then it would be plausible to expect that the DW contrast should change
247 when the magnetic easy axis is rotated, i.e., the zero-contrast angle θ_0 should be offset
248 by 90° between $\text{Co}(2)/[\text{Ni}(2)/\text{Co}(1)]_2/\text{W}(110)$ and $[\text{Co}(1)/\text{Ni}(3)]_2/\text{W}(110)$ as a result of
249 orthogonal magnetization orientations in the two systems. However, the experimental
250 values of θ_0 in the two systems are very close, which indicates that the observed DW
251 contrast may have an extrinsic origin related to the SPLEEM instrumentation.

252 Searching for a possible extrinsic origin of the unusual DW contrast, we carefully
253 explored the dependence of this DW contrast on changing the electron energy to values
254 V_s slightly different from V_{s0} . In Figs. 5(a) and 5(c), SPLEEM images of the
255 $\text{Co}(2)/[\text{Ni}(2)/\text{Co}(1)]_2/\text{W}(110)$ sample at $V_s=5.2$ V and 5.3 V, respectively, clearly show
256 the magnetic domains with opposite contrast. In addition the image at $V_{s0}=5.25$ V
257 shows the DW contrast. Careful inspection reveals that the contrast at the left domain
258 boundary in both Figs. 5(a) and 5(c) is slightly brighter than that at the right domain
259 boundary. This contrast difference can be clearly identified in the line profiles shown
260 in Fig. 5(e), where line profiles of the SPLEEM images at 5.2 V and 5.3 V show small
261 peaks at left DW and small dips at right DW, as indicated by arrows in Fig. 5(e).
262 Inspecting reversed domain contrast above and below V_{s0} , in combination with non-
263 reversing contrast peaks at the domain boundaries, stimulated our hunch to consider the
264 image formed in a pixel-by-pixel sum of the two SPLEEM images acquired at 5.2 V
265 and 5.3 V, just below and above the zero-asymmetry electron energy. The result of
266 summing the images is shown in Fig. 5(d), which looks strikingly similar to the
267 SPLEEM image measured at 5.25 V with zero domain contrast, shown in Fig. 5(c).

268 Similar image summation operation can also be done in the SPLEEM measurements
269 of the $\text{Co}/\text{W}(110)$ system. As shown in Fig. 1, the domain contrast is maximum at 4.5V
270 and 7.5 V, but minimum at 6.2V. If the SPLEEM images acquired at 4.5V and 6.2 V
271 are added together, the resultant image reproduced in Fig. 6(a) shows contrast along

272 DWs that appears very similar to the DW contrast observed in the SPLEEM image
273 measured at 4.9 V [Fig. 1(e)]. Here in order to remove the unbalanced domain contrast
274 at 4.5 V and 7.5 V, the domain contrast in the 4.5 V and 7.5 V images was normalized
275 to 1 before the summation operation. Likewise, summation of images acquired at 6.2 V
276 and 7.5 V results in the image shown in Fig. 6(b), which is similar to the SPLEEM
277 image acquired at 6.9 V [Fig. 1(f)], and reproduces the observed DW contrast. This
278 image summing operation can also be done for images recorded with the electron
279 polarization perpendicular to the domain magnetization, $m \perp \sigma$, and the resultant
280 summed images shown in Figs. 6(c) and 6(d) are very similar to the SPLEEM images
281 shown in Figs. 2(c) and 2(d), reproducing the alternating black and white contrast across
282 the DWs.

283

284 **4. Discussions**

285 Figures 1-4 show that the observed DW contrast is independent of the
286 magnetization orientation within the domains, and independent of the rotation sense of
287 the DW spin texture, which suggests that this DW contrast does not originate from the
288 magnetic properties. Moreover, the summation operations shown in Figs. 5 and 6 can
289 reproduce the DW contrast observed in the SPLEEM images measured at V_{s0} . This
290 observation suggests a possible extrinsic origin related to the SPLEEM instrumentation,
291 as we discuss below.

292 As shown by the reflectivity spectra in Fig. 1(a) and Fig. 3(a), the electron
293 reflectivity quickly drops for the electron energy above the workfunction within a
294 certain energy range, and the energy width of this dropoff corresponds to the energy
295 width of incident electron beam. The energy width of the spin polarized beam used in
296 these measurements is estimated to be ~ 0.3 eV, this energy distribution is sketched
297 schematically in Fig. 7(a). While the SPLEEM measurement is performed at V_{s0} , half
298 of the electrons with lower energy will produce an image with positive domain contrast,
299 while the other half of the electrons with higher energy will produce an image with the
300 opposite domain contrast, as sketched in Fig. 7(b), left- and middle panels. If these two
301 images, acquired with energy below and above V_{s0} , are slightly displaced from each

302 other, then contrast can appear at domain walls. SPLEEM images correspond to
303 averages of all the electrons: as shown in the right panel of Fig. 7(b), representing the
304 pixel-by-pixel average of the left and middle panels, under conditions of energy
305 dependent image shift the magnetic domain contrast vanishes in all image areas, except
306 in close proximity to the DWs. Due to the displacement between images resulting from
307 different tails in the beam energy distribution, bright and dark bands outline the DW,
308 and this effect vanishes only in sections where the DW is oriented parallel to the
309 horizontal image shift. The DW contrast at V_{s0} for the incident electron spin
310 perpendicular to the domain magnetization ($\mathbf{m} \perp \boldsymbol{\sigma}$) shown in Fig. 2 can also be explained
311 by the energy-dependent image shifting effect. Under this measurement condition, only
312 the DWs have the magnetic contrast, and the energy-dependent image shifting around
313 V_{s0} can result in the oscillating white and black contrast cross the DWs.

314 In this picture, it is clear that, if the asymmetry spectrum has the opposite slope as
315 that shown in Fig. 7(a), then the domain contrast should also be opposite to that shown
316 in Fig. 7(b). This is consistent with the observation of the opposite DW contrast at 4.9
317 V and 6.9 V in the Co/W(110) system (Fig. 1), which can be attributed to the opposite
318 slope at these two energies in the asymmetry spectrum. Likewise, in Fig. 4(c) and 4(d)
319 the reversed DW contrast can be attributed to the opposite slope in the corresponding
320 asymmetry spectra shown in Fig. 4(a) and 4(b).

321 Based on this picture, we can quantitatively reproduce the observed DW contrast
322 through simulations. We consider a circular domain with the diameter of 3.6 μm ,
323 surrounded by a DW with a width of 150 nm, similar to magnetic domain structures
324 imaged in Fig. 1. As shown in Fig. 7(a), we model the incident electron beam as having
325 a Gaussian energy distribution with the half-peak width of $\Delta V = 0.3$ V. With
326 approximately linear dependence of the asymmetry on the electron energy (4.9% per
327 electron volts derived from the asymmetry spectrum in Fig. 1), we can compute a
328 weighted sum of all domain images across this energy range, accounting for image
329 weight following the Gaussian distribution. To include the energy-dependent image
330 shift, we assumed that images are shifted along the horizontal direction by 430 nm/V.
331 The resulting simulated image shows bright contrast in the left DW and dark contrast

332 in the right DW. The amplitude of the simulated DW contrast is about 0.49% at V_{s0} .
333 Considering that the measured DW contrast is $\sim 0.29\%$ at 4.9 V and $\sim 0.50\%$ at 6.9 V,
334 our simulation quantitatively reproduces the measured DW contrast, which further
335 supports our interpretation that the observed DW contrast at V_{s0} is likely due to beam
336 energy spread combined with energy-dependent image shift.

337 The suggested energy-dependent image shift can be measured by modulating
338 the electron energy in the SPLEEM. To this end a voltage source was connected to the
339 cathode in the SPLEEM electron gun to measure how much the image position can be
340 influenced by modulating the electron beam energy without changing the imaging
341 condition in the imaging column. This experiment was performed on a high-contrast
342 sample of Pd(0.15ML)/Ru(0001). Fig. 7(c) shows that the LEEM image of this Pd/Ru
343 surface contains Pd nano-dots with bright contrast [36], and Fig. 7(d) shows that the
344 image is shifted by ~ 150 nm to the left, as indicated by the red outlines in Fig. 7(c) and
345 7(d), when the cathode potential is raised by 0.35 V. For reference, the orange circle
346 highlights a permanent defect on the phosphor screen. Thus the magnitude of the
347 energy-dependent image shift is ~ 430 nm/V, as used in our simulation described above.
348 The direction of this observed energy dependent image shift coincides with the beam-
349 deflection plane of the magnetic beam separation prism, approximately 10° with respect
350 to the image horizontal direction of our CCD camera. As a final test, we reproduced the
351 observed DW contrast by modulating the electron beam energy by applying a 1000 Hz
352 square wave voltage to the cathode. SPLEEM image acquisition time is usually in the
353 range of second, thus the electron energy modulation at much higher frequency
354 essentially simulates a controlled increase of the energy width of the beam. Figs. 7(e)
355 and 7(f) show the measured DW images at $V_{s0} \sim 4.9$ V under energy modulation with
356 the amplitudes of 0.2 V and 0.4 V respectively. A clearer DW contrast can be identified
357 in Fig. 7(f) for the higher amplitude of electron energy modulation, showing that
358 increased beam energy broadening induces stronger DW contrast at V_{s0} where the
359 contrast between homogeneously magnetized domains vanishes.

360

361 **5. Conclusion**

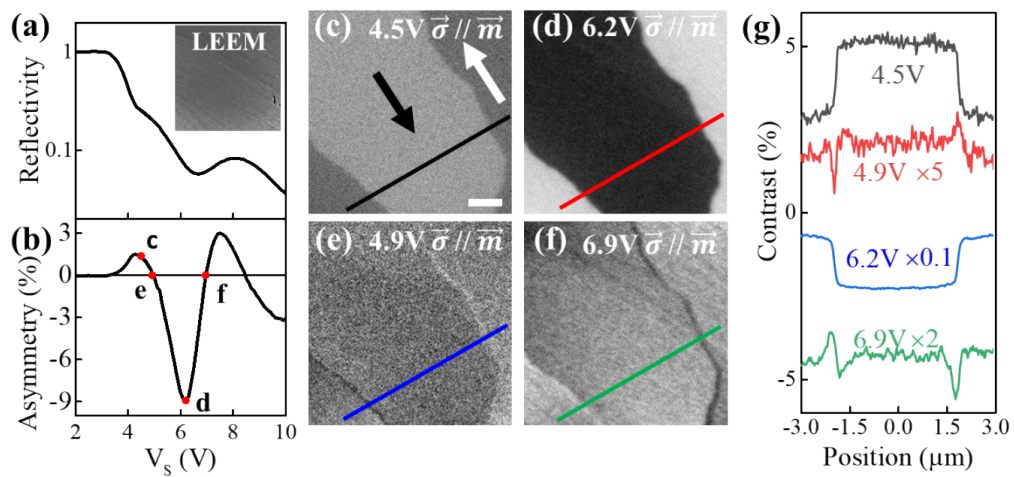
362 In summary, we observed unusual DW contrast in SPLEEM measurement on
363 different magnetic films. This DW contrast appears only at electron energy where the
364 contrast between homogeneously magnetized domains vanishes. This contrast is related
365 to the slope of the spin asymmetry spectrum at such energies, and is independent of the
366 spin rotation sense inside the DWs. Our control experiments indicate that this new DW
367 contrast is attributed to the energy width of the incident electron beam and energy-
368 dependent image shifts resulting from the dispersive properties of the electron optics
369 used in the SPLEEM instrument. Although the energy spread of the electron beam can
370 be neglected in the interpretation of most SPLEEM measurements, dependence of the
371 contrast observed here on the slope of spin asymmetry spectra suggests that the beam
372 energy broadening may introduce effects that must be taken into account in some
373 circumstances. This DW contrast might not play a significant role in the SPLEEM
374 images acquired at beam energy with strong domain contrast, but it may influence spin
375 texture measurements in experiments investigating conditions with weak spin contrast.
376 The observed DW contrast is proportional to the energy-dependent slope of the spin
377 asymmetry spectrum, thus it doesn't influence the interpretation of earlier DW studies
378 using SPLEEM with the electron energy at the maxim domain contrast. However, our
379 work suggests that interpretations of complex SPLEEM images with non-collinear
380 domain structures may benefit from careful analysis.

381

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394 Fig. 1 (a)-(b) Reflectivity and spin asymmetry spectra of Co(4ML)/W(110). Inset is a

395 LEEM image of this sample taken with $V_s=5$ V. The red dots in (b) indicate the electron

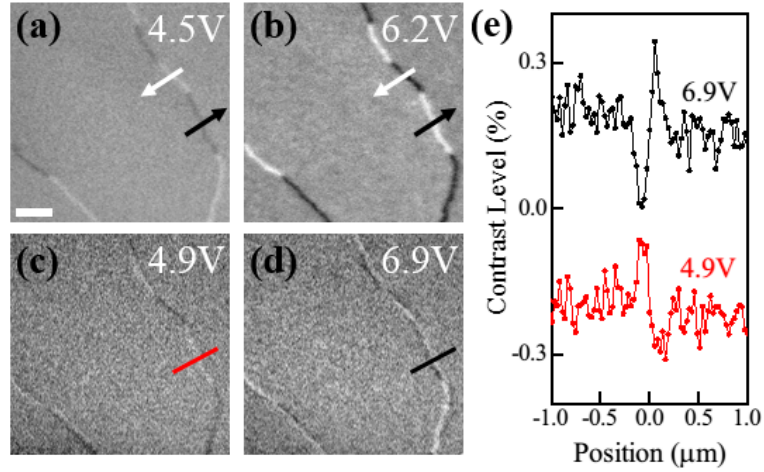
396 energies V_s used for the SPLEEM images in (c-f). (c)-(f) SPLEEM images of the same

397 position at different V_s (4.5 V, 4.9 V, 6.2 V, 6.9 V) for the electron polarization $\vec{\sigma}$

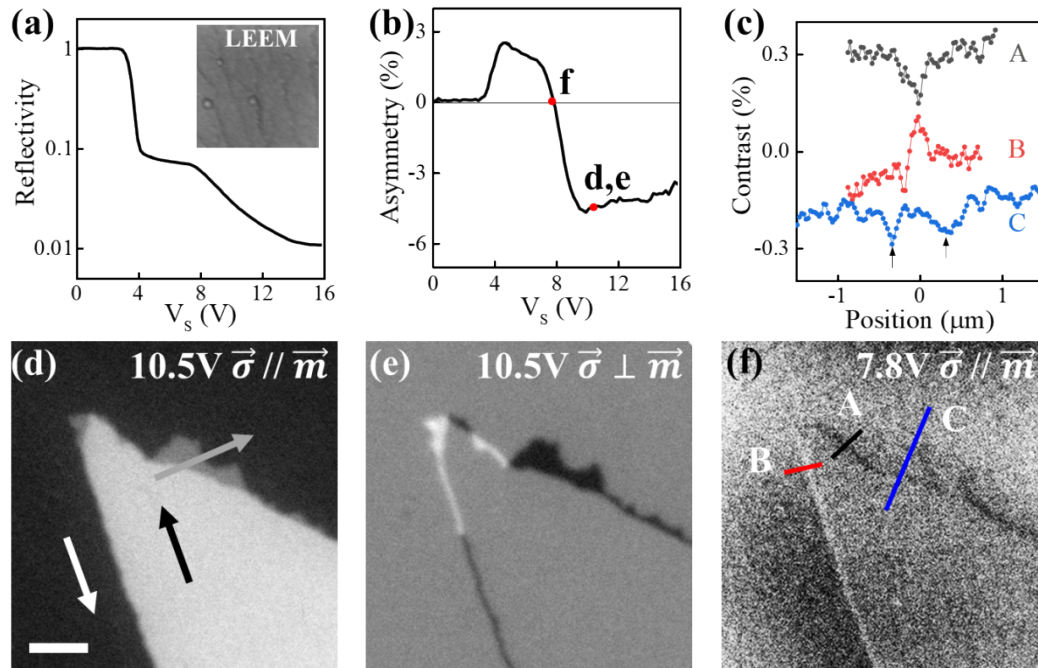
398 parallel to the magnetization \vec{m} in the domains ($m//\sigma$). The scale bar in (c) is 1 μm .

399 (g) Line profiles from (c)-(f) in the area marked with the lines in corresponding color..

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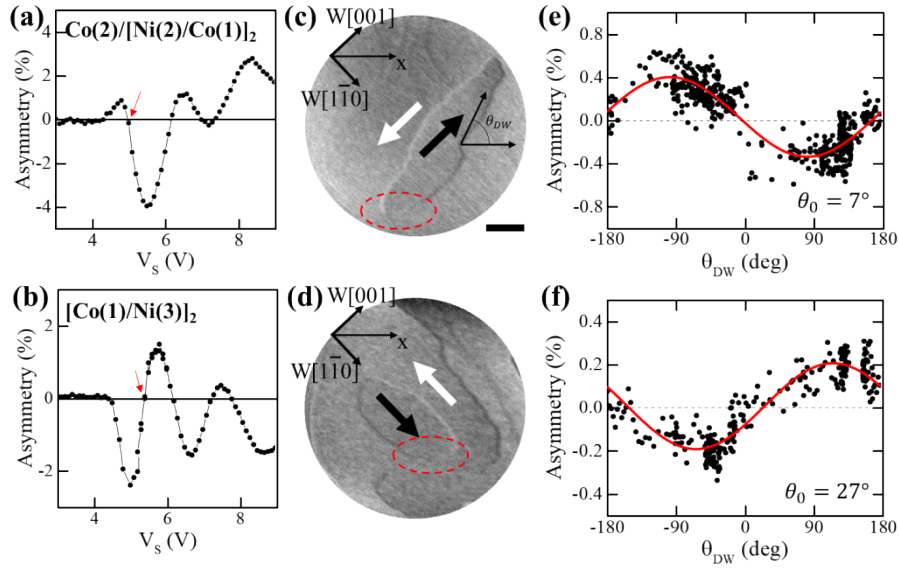
401
 402 Fig. 2 (a)-(d) SPLEEM image of the same position as Fig. 1 at different V_S (4.5 V, 4.9
 403 V, 6.2 V, 6.9 V) with electron polarization $\vec{\sigma}$ perpendicular to the magnetization \vec{m}
 404 in the domains ($m \perp \sigma$). The scale bar in (a) is 1 μm . (e) Line profiles from the
 405 corresponding-color lines cross the DWs in (c) and (d), which demonstrate the
 406 alternating white and dark contrast across the DWs.
 407



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409 Fig. 3 (a) Reflectivity spectrum and (b) Spin asymmetry spectrum of Co(15
 410 ML)/Cu(001). Inset in (a) is the LEEM image of this sample with $V_s=5$ V. The red dots
 411 in (b) indicate the electron energies V_s used for the SPLEEM images in (d-f). (c) Line
 412 profiles from the marked positions in (f). (d)-(f) SPLEEM images of the same position
 413 with different combinations of V_s and the electron polarization direction: (d) $V_s=10.5$
 414 V, $\vec{\sigma} // \vec{m}$, (e) $V_s=10.5$ V, $\vec{\sigma} \perp \vec{m}$, and (f) $V_s=7.8$ V, $\vec{\sigma} // \vec{m}$. The scale bar in (d)
 415 is 1 μm . Arrows in (d) indicate magnetization directions.

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418 Fig. 4 Asymmetry spectra of (a) $\text{Co}(2)/[\text{Ni}(2)/\text{Co}(1)]_2/\text{W}(110)$ and (b)

419 $[\text{Co}(1)/\text{Ni}(3)]_2/\text{W}(110)$. SPLEEM images in (c) of $\text{Co}(2)/[\text{Ni}(2)/\text{Co}(1)]_2/\text{W}(110)$ with

420 $V_s=5.25$ V, and (d) of $[\text{Co}(1)/\text{Ni}(3)]_2/\text{W}(110)$ with $V_s=5.34$ V. The scale bar in (c) is 2

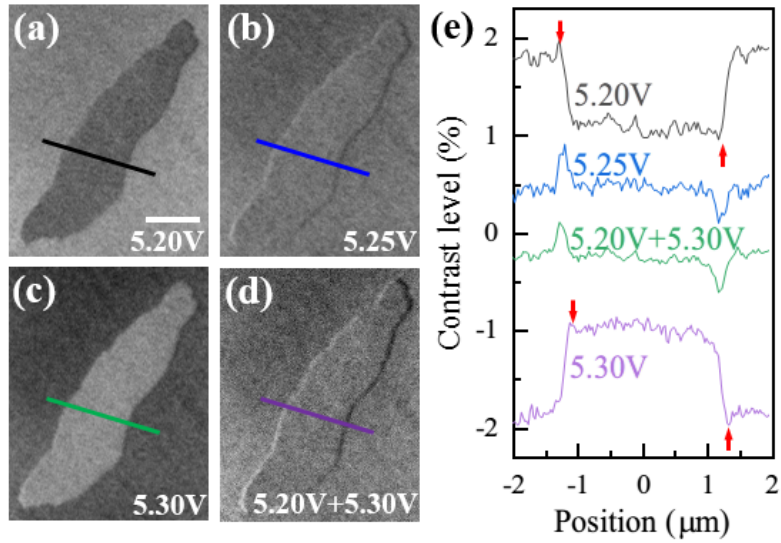
421 μm . The red circles in (c) and (d) mark the DW section where the unusual DW contrast

422 vanishes. (e)-(f) Contrast inside the DWs in (c) and (d) as a function of DW orientation

423 θ_{DW} , as defined in (c). The red lines at fitting curves with the function $A_0\sin(\theta_{\text{DW}}-\theta_0)$.

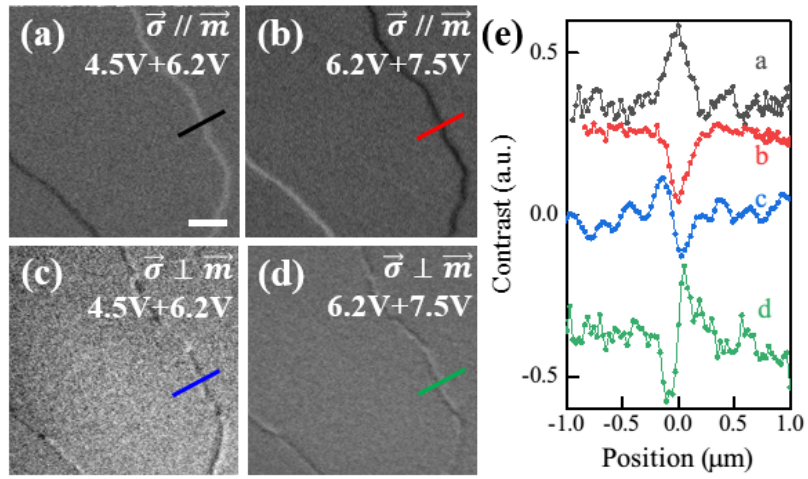
424 The best-fit values of θ_0 are indicated inside the figures.

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427 Fig. 5 (a)-(c) SPLEEM images of $\text{Co}(2)/[\text{Ni}(2)/\text{Co}(1)]_2/\text{W}(110)$ with V_s of 5.2 V, 5.25
 428 V and 5.3 V respectively. (d) Pixel-by-pixel averaged image of the SPLEEM images at
 429 5.2 V in (a) and 5.3 V in (c), which is similar to the measured SPLEEM image at 5.25
 430 V in (b). The scale bar in (a) is 2 μm . (e) Line profiles across the corresponding-color
 431 lines in (a)-(d). Red arrows indicate small peaks and dips in the contrast, see main text.
 432

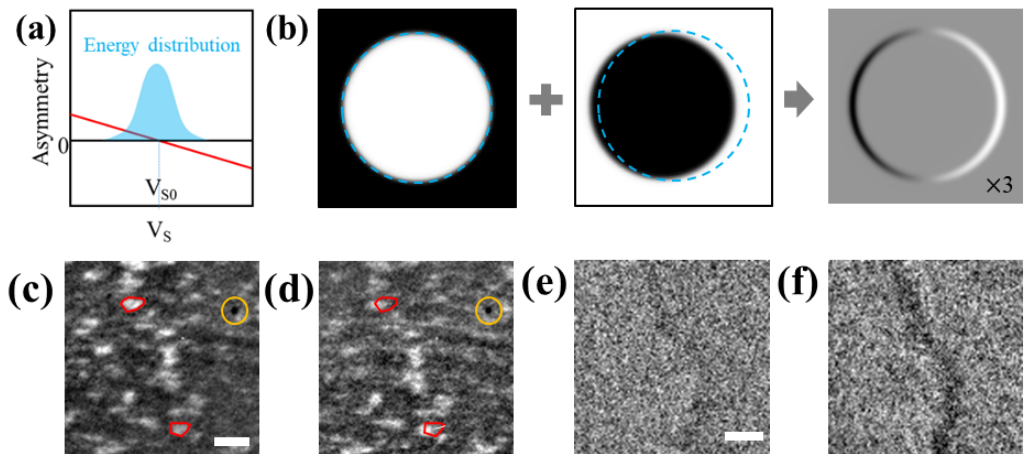


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434 Fig. 6 (a)-(d) Averaged SPLEEM images of Co(4 ML)/W(110). The values of V_s and
 435 the spin polarization orientation are marked in the figures. The scale bar in (a) is 1 μm .

436 (e) Line profiles from the corresponding-color lines in (a)-(d).

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439 Fig. 7 . (a) Schematic drawing of spin asymmetry spectrum (red curve) near V_{s0} , and
 440 the distribution of electron energy (blue area). (b) Schematic drawing of the contrast
 441 simulation of a circular magnetic domain, including the energy dependent image
 442 displacement due to dispersion in the beam separation prism array in our SPLEEM
 443 instrument. Image at higher electron energy (middle) is displaced slightly to the left
 444 with respect to the image at lower energy (left). Averaging these two images (right)
 445 results significant contrast on domain boundary, and the simulated DW contrast was
 446 enhanced by a factor of 3, which means the simulated DW contrast is about one third
 447 of the domain contrast at $V_{s0} \pm 0.3$ V (see main text). (c) LEEM image of Pd(0.15
 448 ML)/Ru(0001), bright areas are Pd islands, dark areas are bare Ru surface. (d) LEEM
 449 image obtained with electron gun cathode potential raised by additional 0.35 V,
 450 imaging conditions otherwise identical to (c). Red outlines highlight the image shift in
 451 panel c and d, for clarity orange circle highlight a defect on screen which does not
 452 move. (e) and (f), SPLEEM images of Co(4 ML)/W(110) at 4.9 V while applying a
 453 1000 Hz square wave voltage the cathode with the amplitude of (e) 0.2 V and (f) 0.4 V
 454 respectively. Scale bars in (c) and (e) are 500 nm.

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