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Domain wall dynamics in two-dimensional van der Waals ferromagnets

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Domain wall motion is in the core of many information technologies ranging from storage[1], processing[2], sensing[3] up to novel racetrack memory architectures[4]. The finding of magnetism

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[21] in two-dimensional (2D) van der Waals (vdW) materials[5–8] has offered a new frontier for the

[22] exploration and understanding of domain walls at the limit of few atom-thick layers. How-

[23] ever, to use 2D vdW magnets for building spintronics nanodevices such as domain-wall based

[24] logic[9–11], it is required to gain control of their domain wall dynamics by external driving

[25] forces such as spin-polarized currents or magnetic fields which has so far been elusive. Here

[26] we show that electric currents as well as magnetic fields can efficiently move domain walls in

[27] the recently discovered 2D vdW magnets CrI_3 and $CrBr_3$ at low temperatures, and robust

[28] down to monolayer. We realize field- and current-driven domain wall motion with veloci-

[29] ties up to 1020 m s^{-1} which are comparable to the state-of-the-art materials for domain-wall

[30] based applications[12–16]. Domain walls keep their coherence driven by the spin-transfer torque

[31] induced by the current and magnetic field up to large values of about 12×10^9 A cm^{-2} and

[32] 5 T, respectively. For larger magnitudes of current or field, a transition to a hydrodynamic

[33] spin-liquid regime is observed with the emission of a periodic train of spin-wave solitons

[34] with modulational instability[17]. The emitted waveform achieves terahertz (THz) frequency

[35] in a wide range of field and current densities which opens up perspectives for reconfigurable

[36] magnonic devices. Moreover, we found that these spin-waves can transport spin angular mo-

[37] mentum through the layers over distances as long as 10 μm without losses for the transport

[38] of spin information. Our results push the boundary of what is currently known about the

[39] dynamics of domain walls in 2D vdW ferromagnets and unveil strategies to design ultrathin,

[40] high-speed and high-frequency spintronic devices.

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Spin-based applications have been broadly explored for high-performance solid-state data storage technologies[18,19]. A promising strategy is to encode bits in magnetic domains walls which can be controlled via applied magnetic fields and spin-polarized currents[4,9]. Several approaches for domain wall-based memory devices with a focus on the domain wall displacement have been developed. From a field-controlled shift register[9,20] up to the non-volatile multi-turn sensors[21], which are commercially available[22], these emerging technologies provide a new horizon for advanced materials to be explored. With the discovery of magnetism in vdW layered compounds[5–7] and proof-of-concept devices[8,23–25] already showing outstanding progress toward applications, one of the main challenges is the integration of 2D magnets in domain wall microelectronic platforms.

Due to scaling reasons and high-density requirements[26], competitive devices should have narrow domain walls (ideally in the range of $1-10$ nm), use out-of-plane magnetic anisotropy materials and, in particular for memory devices, develop domain wall velocities of the order of ~ 100 m s^{-1} to achieve rapid operating speed on increasing areal density[4,27]. Sufficiently low current densities ($\sim 10^6 - 10^8$ A cm^{-2}) are also needed to guarantee low power consumption and to avoid damage due to Joule heating. However, metallic layers inherently suffer with energy dissipation via the conduction electrons[2] which make magnetic insulators better suited for domain wall applications. There have been a few reports on the creation of different interfaces using magnetic vdW materials[28,29] and heavy metal overlayers (e.g. Pt, Ta) where spin-transfer torque arising from charge-to-spin current conversion can effectively switch the magnetization from one state to another. Nevertheless, it is unknown how domain walls behave intrinsically in 2D magnets as current and fields are applied directly into the systems, and how efficient such layered structures

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62 would be in domain-wall based functional devices.

63   Here we demonstrate domain-wall motion in monolayer CrI_3 and $CrBr_3$ by spin polarized

64 currents and magnetic fields in a broad range of densities ($10^7 - 10^9$ A cm^{-2}) and field ($0.001-2$

65 T). Both dynamics occur over hybrid domain walls with Néel-Bloch characteristics with a domain

66 wall width of about ~ 5.30 nm. The spin-torque induced by electric currents and magnetic fields

67 generates a rapid rotation of the magnetization at the domain wall, hence triggering its transla-

68 tional motion at high velocities. We record speeds of up 1020 m s^{-1} and 100 m s^{-1} with current

69 and field, respectively, while keeping the coherence of the wall profile through a steady motion.

70 As the domain wall velocity approaches the maximum spin-wave group velocity, the domain wall

71 motion start to exhibit non-linear effects with the emission of magnons at terahertz (THz) fre-

72 quencies following a modulational instability behaviour. Even though no conduction electrons are

73 present in the medium due to the insulating characteristics of CrI_3 and $CrBr_3$, spin-information can

74 be transported over distances as long as 10 μm which sets a new paradigm for spin-wave-based

75 technologies at the ultrathin limit.

76   Our starting point is the investigation of the magnetic domain structures in monolayer $CrBr_3$

77 and their dynamic evolution under different temperatures and magnetic fields. The domain wall

78 structure of CrI_3 and its cooling dynamics have recently been reported using similar theoretical

79 framework[30]. We use a large square flake of 0.4 μm $\times$ 0.4 μm to represent a system able to

80 be measured using imagining techniques, i.e. nitrogen-vacancy scanning magnetometry[31]. We

81 describe the interactions using the following spin Hamiltonian:

$$\mathcal{H} = -\sum_{ij} J_{ij}(\mathbf{S}_i \cdot \mathbf{S}_j) - \sum_{ij} \lambda_{ij} S_i^z S_j^z - \sum_i D_i(\mathbf{S}_i \cdot \mathbf{e}_i)^2 - \sum_{ij} K_{ij}(\mathbf{S}_i \cdot \mathbf{S}_j)^2 - \sum_i \mu_i \mathbf{S}_i \cdot \left(\mathbf{B}_i + \mathbf{B}_i^{\mathrm{dp}}\right) \quad (1)$$

82   where $\mathbf{S}_i$ and $\mathbf{S}_j$ are the localized magnetic moments on Cr atomic sites i and j which are coupled

83   by pair-wise exchange interactions. J_{ij} and λ_{ij} are the isotropic and anisotropic bilinear (BL) ex-

84   changes, respectively, and D_i is the single ion magnetic anisotropy. $\mathbf{B}_i$ and $\mathbf{B}_i^{\mathrm{dp}}$ represent external

85   and dipole magnetic field sources respectively. Eq. 1 was previously found to describe accurately

86   the magnetic properties of several 2D magnets[30,32] including CrI$_3$ and CrBr$_3$. Comparisons with

87   other spin Hamiltonians such as Kitaev, bilinear Heisenberg, Ising, and Dzyloshinskii-Moriya in-

88   teractions, were undertaken in refs.[30,32] as well as in their Supplementary Information files. We

89   take into account up to third nearest neighbors for J_{ij} and λ_{ij} in the description of CrBr$_3$ in Eq.1.

90   All the parameters in Eq.1 are extracted using highly accurate non-collinear ab initio simulations

91   taking into account spin-orbit coupling and Hubbard-U corrected density functional theory as de-

92   scribed in Ref.[30,32]. We ensure that fine numerical convergence within $10^{-6} - 10^{-8}$ eV is achieved

93   in each computed parameter. We also compared the exchange parameters used in our work with

94   those collected from the literature[33]. In the context of the materials studied in our study (monolayer

95   CrI$_3$ and CrBr$_3$), Supplementary Figure 1 clearly shows that our exchange magnitudes[32] reproduce

96   with high accuracy the critical temperatures measured. Moreover, Eq.1 provides an accurate de-

97   scription of the magnon dispersion extracted from inelastic neutron scattering measurements on

98   CrI$_3$[34] as shown in ref.[32]. These results provide a firm background for the modeling of the domain

99   wall dynamics as described here.

100   The fourth term in Eq.1 represents the biquadratic (BQ) exchange which involves the hopping

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101 of two or more electrons between two adjacent sites[32]. Its strength is given by the constant K_{ij},

102 which is the simplest and most natural form of non-Heisenberg coupling. It has recently been found

103 that several 2D vdW magnets develop substantial BQ exchange in their magnetic properties which

104 is critical to quantitatively describe important features such as Curie temperatures, thermal stability,

105 magnon spectra and non-trivial topological spin textures[30, 32, 35, 36]. The magnitude of K_{ij} for $CrBr_3$

106 is 0.22 meV which is smaller than the BL exchange for the first-nearest neighbours ($J_1 = 1.66$ meV)

107 but still sizeable to be disregarded[32]. In our implementation the BQ exchange is quite general and

108 can be applied to any pair-wise exchange interaction of arbitrary range. Each system is evolved by

109 solving the atomistic Landau-Lifshitz-Gilbert equation with a Gilbert damping $\lambda_G = 0.1$ to ensure

110 better computational efficiency and numerical stability of the time integration. The value of λ_G is

111 substantially higher than that (2×10^{-3}) recently measured for a parent compound[37] (i.e. $CrCl_3$)

112 which a similar magnitude would be expected for CrI_3 and $CrBr_3$. The large λ_G also accounts

113 for the possibility of free-electron doping to raise the conductivity of the system due to different

114 processes (i.e. dopants[38], electric bias[39]). Due to the inclusion of dipole-dipole interactions we

115 explicitly include non-local damping effects in our simulations such as due to domain walls or

116 different magnetic textures. Hence, our spin-dynamics simulations include an accurate description

117 of the spin-interactions at the atomistic level (2−20 Å) in a multiscale framework at the lab scale

118 (a few μm's).

119 Monolayer $CrBr_3$ is thermally equilibrated above 40 K and then linearly cooled to 0 K in

120 a total simulated time of 4.0 ns at different magnetic fields (Figure 1 and Supplementary Movies

121 S1-S4). The time evolution of the out-of-plane magnetization M_z is utilised to study the nucleation

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of the magnetic domains as it determines the easy-axis throughout the layer. For zero-field cooling (0 mT) magnetic domains are formed for temperatures below 20 K with the free motion of the domain walls as the temperature drops (Fig. 1**a-e**). As the system reaches 0 K a continuous time evolution of the domain structures remains which results in a homogenous magnetization over the entire crystal (Supplementary Movie S1). Even though thermal fluctuations as well as their contributions to the energy of the system are inexistent at 0 K, other terms, such as dipolar fields, exchange interactions and magnetic anisotropy, are still present, further contributing to the modification of the magnetic domains. Once magnetic fields are applied to the nanosheet (Fig. 1**f-t** and Supplementary Movies S2-S4) this nucleation-type mechanism is accelerated with a rapid reversal in the sign of the magnetization M_z. Indeed, a monodomain feature is observed even in fields far below the coercivity[40] (10 mT at 5 K) for $CrBr_3$. In a scenario where defects or pinning sites are not present in monolayer $CrBr_3$, magnetic domains tend to be meta-stable with a homogeneous magnetisation throughout the surface. This is in sound agreement with recent NV-center scanning magnetometry measurements[31] on the magnetic domain evolution of $CrBr_3$ indicating that pinning effects are the dominant coercivity mechanisms.

Strikingly, the interplay between domain metastability and high magnetic anisotropy of $CrBr_3$ give additional characteristics to the domain walls (Figure 2**a-b**). We observe that as the spins vary orientation from one magnetic domain to another the wall profile assumes components of the magnetization along different in-plane directions ($M_{x,y}$) relative to the easy-axis (M_z) (Fig. 2**c-d**). In-plane magnetization M_x displays a variation larger than that for M_y which is mainly noticed at the core of the domain wall within $1-2.5$ nm. These features indicate a domain wall with

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143 hybrid characteristics of Bloch and Néel type (Fig. 2**e-f**). We can extract the domain wall width

144 $\sigma_{x,y,z}$ by fitting the different components of the magnetization (M_x, M_y, M_z) to standard equations

145 of the form[41]:

$$M_j = \frac{1}{\cosh(\pi(j - j_0)/\sigma_j)}, \quad \text{with} \quad j = x, y \tag{2}$$

$$M_z = \tanh(\pi(z - z_0)/\sigma_z) \tag{3}$$

146 where j_0 and z_0 are the domain wall positions at in-plane and out-of-plane coordinates, respec-

147 tively. The domain wall widths are within the range of $\sigma_{x,y,z} = 5.30 - 5.33$ nm. Materials with

148 high magnetic anisotropy generally stabilise such small domain wall widths but their magnetic do-

149 mains are generally stable after zero-field cooling due to long range dipole interactions. We have

150 checked that the inclusion of dipolar fields in our simulations do not change the results.

151 An intriguing question raised by the hybrid features of the domain walls in $CrBr_3$ and CrI_3[30]

152 is the effect of magnetic fields and electric currents on the motion of domain walls. It is well estab-

153 lished that both driving forces can induce displacement of domain walls in magnetic thin films over

154 different substrates. Nevertheless, the vdW nature of the nanosheet together with the atomic thick-

155 ness ($\approx$0.5 nm) may induce additional phenomena yet to be observed in 2D ferromagnets. Figure

156 3**a-d** shows that either fields or currents can efficiently displace domain walls in layered $CrBr_3$ and

157 CrI_3 magnets. The dynamics is generally initialised with a domain wall at equilibrium position

158 previously thermalized for several nanoseconds for full convergence of the spin orientations (Fig.

159 3**a**). The finite width of the nanowire assists in the formation of the domain walls pinned by the

160 edges. For field-induced domain wall motion, wall velocities up to $v = 70$ m s^{-1} and $v = 98$ m s^{-1}

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161 are recorded for $CrBr_3$ and CrI_3 (Fig. 4**a-b**), respectively. The wall moves steadily on both sheets

162 and can be described as $v = \mu_F(B - B_o)$, where μ_F is the wall mobility and B_o is the onset field

163 required to move the wall from structural defects or pinning[42]. The magnitudes of $\mu = 0.0343$ m

164 s^{-1} mT^{-1} and $B_o = 23.89$ mT for $CrBr_3$ and $\mu = 0.018$ m s^{-1} mT^{-1} and $B_o = 13.33$ mT for CrI_3

165 indicate that domain walls move relatively slower than those in metallic ultrathin Pt/Co/Pt films[43]

166 or thick magnetic insulator TmIG/Pt interfaces[14] but with relatively similar onset field B_o. The

167 domain wall moves steadily on both sheets at fields below critical magnitudes (B_c) of $B_c = 1.19$

168 T and $B_c = 4.78$ T for $CrBr_3$ and CrI_3, respectively (Supplementary Movies S5-S6). For values

169 beyond B_c different spin distributions start nucleating ahead of the domain-wall (Supplementary

170 Movie S7-S8) and induce its collapse within a few hundreds of picoseconds after the motion ini-

171 tiated. These spin features appear from the edges of the layers and rapidly move into the bulk of

172 the ribbon. This behaviour is analogous to that observed in Pt/Co/AlO$_x$ ultrathin microstructures[44]

173 and highlighted the importance of edges on the magnetic properties of 2D magnets in device in-

174 tegration. A preference for unreconstructed edges or dangling bonds rather than on zig-zag edges

175 (Fig. 3**b** at 1.50 T) has been observed in the nucleation of reversed domains during the wall dy-

176 namics. The local variation of the exchange interactions at dangling-bonds with spins being less

177 restricted to change their directions with an external field than at zig-zag edges is one of the main

178 ingredients for this behaviour. This observation shall trigger further experimental studies of the

179 atomic structure at the edges of the monolayer magnets via scanning probe microscopies[45,46] and

180 their connection to the device performance.

181   In the case of current-driven domain wall motion, we state that such behaviour can be ac-

182 complished in two ways. First, by using an adjacent conducting layer as recently demonstrated for

183 insulating $Tm_3Fe_5O_{12}$ on top of a Pt substrate[14]. Charge current will flow in the heavy metal gen-

184 erating spin currents that exert a spin-transfer torque (STT) on the ferromagnetic material. Second,

185 by doping the CrX_3 (X=Br, I) magnets in order to generate significant carriers in the host[27,47]. Such

186 strategies are well established and would provide a feasible platform to confirm our predictions.

187 Indeed, we recorded domain wall velocities up to 530 m s^{-1} and 1020 m s^{-1} for monolayer $CrBr_3$

188 and CrI_3, respectively, under applied currents (Fig. 4**c-d**). These velocities are higher than those

189 observed in a wide range of systems including skyrmions (100 m s^{-1})[13], ferromagnetic semicon-

190 ductor (Ga, Mn)As (22 m s^{-1})[47], synthetic antiferromagnets (750 m s^{-1})[12], metallic layers (380 m

191 s^{-1})[48], Pt/CoFe/MgO and Co/Ni/Co interfaces ($\sim$10 m s^{-1}, 400 m s^{-1})[16,49], and insulating oxides

192 (400 m s^{-1})[14]. It is worthwhile highlighting that low current densities within the experimental

193 range of 10^{-8} A cm^{-2} (insets in Fig. 4**c-d**) already resulted in domain wall speeds (i.e. 100 m s^{-1})

194 similarly as those achieved in established racetrack platforms with more complex materials[4]. The

195 domain walls displace freely as a function of the current density j in a viscous flow motion (Supple-

196 mentary Movies S9-S10). The high velocities predicted for $CrBr_3$ and CrI_3 are only limited by the

197 spin-wave group velocity v_g intrinsic to the systems. By using a spin-wave theory for 2D magnetic

198 materials[32], which includes biquadratic exchange and Dzyaloshinskii-Moriya interactions (DMI),

199 we estimate from the magnon dispersion (Supplementary Figure 2) maximum values of v_g within

200 the range of $584-891$ m s^{-1} for $CrBr_3$ and $1326-6400$ m s^{-1} for CrI_3 (Supplementary Figure

201 3). These values are close to those computed from the atomistic simulations (530 m s^{-1} and 1020

202 m s^{-1} for $CrBr_3$ and CrI_3, respectively) although slightly larger. This is due to the damping of

10

203 the domain wall motion by the emission of spin-waves[50] not taken into account into the model as

204 commented in the following.

The steady displacement of the domain walls occurs up to a certain threshold (j_w) defined

as the Walker breakdown[42]. For values below j_w, the wall dynamics can be well modelled by a

simple 1D model[51,52] through a linear dependence of v and j:

$$v = \mu_I j \tag{4}$$

205 where μ_I is the current-driven domain-wall mobility which gives $\mu_I = 0.12 \times 10^{-10}$ m^3 A^{-1}s^{-1}

206 and $\mu_I = 0.10 \times 10^{-10}$ m^3 A^{-1}s^{-1} for monolayer CrBr$_3$ and CrI$_3$, respectively. These values

207 are of the same order of magnitude as those recorded on thin metallic interfaces[49] or permalloy

208 nanowires[53] which have been studied more intensively. For values near j_w or above, the spins

209 at the domain wall precesses inducing disruption of the wall at longer times with the nucleation

210 of magnetic domains with a different polarisation ahead of the displacement of the domain-wall

211 (Fig. 3**c** at 4.35×10^9 A cm^{-2}). A close look at these features (Fig. 3**d**) indicates that they

212 may start from both edges although dangling-bonds may induce more curvature to the domain

213 wall since a dragging on the motion is observed generating retardation relative to the zig-zag edge.

214 We can model qualitatively the dynamics at $j > j_w$ for both CrBr$_3$ and CrI$_3$ via the time average

215 velocity of the domain wall[51,52]. This suggests that the domain-wall dynamics in 2D magnets is

216 similar as that in magnetic nanowires which allows faster integration into existing devices[27]. We

217 also observe that as the domain wall moves spin-waves or magnons are emitted (Fig. 4**e-f**). The

218 spin frequencies driven by the field (ω_B) and current (ω_J) are in the terahertz (THz) regime with

219 maximum magnitudes of $\omega_B^{max} = 0.43$ THz and $\omega_J^{max} = 0.60$ THz for CrBr$_3$ and $\omega_B^{max} = 0.50$ THz

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220   and $\omega_J^{max} = 0.66$ THz for CrI$_3$. These frequencies are in sound agreement with those estimate from

221   the magnon dispersion along different points of the Brillouin zone for both halides (Supplementary

222   Figure 3). We found that the maximum frequencies with the field are close to those at the M$-$point

223   (0.47 THz for CrBr$_3$, and 0.57 THz for CrI$_3$), whereas with the applied current are around the

224   K$-$point (0.63 THz for CrBr$_3$, and 0.64 THz for CrI$_3$). Indeed, the variations of ω_B and ω_J can be

225   well fitted using:

$$\omega_B = \omega_0^B + \alpha_B B \tag{5}$$

$$\omega_J = \omega_0^J + \alpha_J j \tag{6}$$

226   where $\omega_0^B(B = 0)$ and $\omega_0^J(j = 0)$ are the onset frequencies resulting in $\omega_0^B = 0.29$ THz and

227   $\omega_0^J = 0.31$ THz for CrBr$_3$; and $\omega_0^B = 0.25$ THz and $\omega_0^J = 0.43$ THz for CrI$_3$. We also notice

228   that the magnitudes of ω_0^B and ω_0^J may also change due to the inhomogeneous spatial distribution

229   of the emitted spin-waves (Supplementary Movie 11) even though with a similar behaviour with

230   the field and current. The coefficient α_B differs moderately between both halides converging to

231   0.13 THz T^{-1} and 0.05 THz T^{-1} for CrBr$_3$ and CrI$_3$, respectively, with $\alpha_J \approx 0.04$ THz 10^{-9} A

232   cm^{-2} being similar to both systems. On the field-driven spin-wave emission, the domain wall pre-

233   cesses with a frequency near the Larmor frequency $\omega_0 = \gamma B$, where γ is the gyromagnetic ratio,

234   similarly as in nanowires[54]. We noticed that in both layered materials the spin wave frequencies

235   are enclosed between odd- and even-numbered harmonics. That is, CrBr$_3$ is within $4\omega_0$ and $5\omega_0$

236   $(\omega_B - \omega_o^B = 4.81\omega_0)$, and CrI$_3$ is within ω_0 and $2\omega_0$ $(\omega_B - \omega_o^B = 1.70\omega_0)$. This indicates that the

237   spin waves and their overtones can be excited with a combination of fundamental modes which

238   gives additional flexibility for terahertz source of magnetic signal using atomically thin layers. In

12

239 addition, the emission of spin waves electrically-driven can be associated with the Doppler effect

240 on the frequency shift[55]. As the domain wall moves the current changes the spin-waves disper-

241 sion and incidentally causes an effective flow of the magnetic medium. This is reflected by the

242 linear dependence of ω_J on j (Fig. 4f) in agreement with the spin-transfer-torque Doppler shift. A

243 similar approach has been successfully used to study the effect of electric signal into magnons in

244 permalloy strips[56], $Ni_{80}Fe_{20}$ wires[57] and thin films[58].

245   A substantially different phenomenon is observed when the domain wall is subject to large

246 current densities, $j > 10^{10}$ A cm^{-2}, with the appearance of spin hydrodynamic effects in $CrBr_3$

247 and CrI_3. The dynamics is illustrated in Figure 5**a-c** where we plot line-cuts of the M_z and M_x

248 components of the magnetisation at width 25 nm and selected times for monolayer CrI_3. We

249 observe that shortly after the electric pulse starting propagating into the system within 18-78 ps

250 the M_x projection exhibits a wave propagating ahead of the domain wall. This amplitude tilts

251 M_z into the plane and increases exponentially (Fig. 5**d**) until M_z is completely suppressed and

252 eventually switches, nucleating a new domain wall. The first period of the wave in M_x or shock

253 wave continues to propagate with no appreciable dissipation and new domain walls are nucleated

254 in its wake with a certain periodicity (Fig. 5**a-c**). This nonlinear phenomenon is indicative of a

255 modulational instability (MI) as observed in fluid dynamics[17,59]. This type of instability occurs in

256 focusing media and describes the exponential growth of waves within a band of wavevectors until

257 non-linear effects favour their spatial localisation into solitons. MI are ubiquitous in dispersive

258 physical problems and have been observed in several systems such as in water waves[60], nonlinear

259 optics [61] and Bose-Einstein condensates[62]. In magnetic materials however MI has been invoked

13

260 so far as a pathway for the nucleation of dissipative droplets in perpendicular magnetic anisotropy

261 (PMA) ferromagnets[63], as well as the spatial localisation of high-energy magnons in PMA ferri-

262 magnetic alloys excited with ultrafast optical pulses[64], and the disintegration of short-wavelength

263 spin-superfluids in planar ferromagnets[65]. In contrast, we find clear evidence of spin hydrodynamic

264 behaviour manifested the onset of MI and its nonlinear dynamics driven by electrical currents in

265 2D magnetic materials. We emphasize that the nonlinear effect observed here is fundamentally

266 different from Walker breakdown and the subsequent precessional regime so far observed in bulk

267 magnets.

268 We also observe that MI is visible already at 20 ps as the wavefront exhibits a transverse

269 structure and can be more generally regarded as a spin shock wave (Fig.6**a-c**). At 200 ps, there are

270 rich transverse features in the nanoribbon, from which we can identify a modulationally unstable

271 region where new domains are established. These domains are bounded by the trailing domain-wall

272 (soliton edge) and the spin shock wave. At 600 ps, the shock wave is already outside our simulation

273 profile and the domain walls continue to propagate further (see Supplementary Movie S12 for the

274 full sequence). The salient features of the MI dynamics can be visualized in Fig. 6**d** by a colour-plot

275 of the M_z component line-cuts as a function of time. After an initial translation of the domain wall,

276 there is a split of behaviours at ≈ 0.01 ns where we can define two characteristic speeds (dashed

277 lines in Fig. 6**d**). The shock wave (SW) (Fig. 6**b**) translates with a speed of $s^+ = 5.77$ km s^{-1} $\pm$

278 0.84 km s^{-1} which is in sound agreement with the maximum group velocity calculated for CrI$_3$

279 (Supplementary Figure 3). The soliton edge exhibits a phase-shift and subsequent translation with

280 a speed of $s_1^- = 1.33$ km s^{-1} ± 0.002 km s^{-1} (Fig. 6**b**). Consequently, the wavelength L of the

14

281 nucleated domains can be extracted from the spectral function at a constant speed, such at s^+

282 (Fig. 6**e**). The obtained frequency of the wavefront or SW is computed as $v_{SW} =$177.5 GHz$\pm$1.25

283 GHz. We can immediately extract L through a simple speed and frequency relationship via $L =$

284 $s^+/v_{SW} =$ 5.77 km s^{-1}/177.5 GHz$= 32.50 \pm 0.23$ nm which is equivalent to a wave-vector of

285 $k_{SW} = 2\pi/L = 0.19$ nm$^{-1}\pm 0.00003$ nm^{-1}. Indeed, the shock-wave speed corresponds to the

286 group velocity of the most unstable magnon with a wave-vector given by k_{SW}.

287   The growing region bounded by the speeds s^+ and s_1^- is reminiscent to the development of

288 dispersive shock waves (DSWs)[17] which is located at the modulationally unstable region high-

289 lighted on Fig.6**b**. DSWs are ordered structures that smoothly connect the wavevectors at the front

290 and soliton edges, and have been fully characterised for planar ferromagnets[66]. However, the MI

291 in our system induces periodic domain-wall nucleation defined by the most unstable wavevec-

292 tor. Therefore, we can regard the scenario of Figs.6**b-c** as an unstable generalization of a DSW.

293 The periodicity of the nucleated domains leads to initially similar domain-wall speeds (parallel

294 features in the wake of the wavefront) that are later perturbed by interactions, annihilation, and

295 nucleation events. These can be potentially related to turbulence, thermodynamic behaviour as a

296 soliton gas[67], or rare events such as rogue waves[60]. At a later time (≈ 0.04 ns), a second char-

297 acteristic domain-wall speed $s_2^- = 0.88$ km s$^{-1}\pm 0.001$ km s^{-1} is observed. This second speed

298 appears to be related to an additional source of MI in the wake of the soliton-edge that opposes the

299 current-driven torque. We can extract the frequency of this domain-wall wake (Fig.6**f**) resulting

300 in $v_{wake} =$6.66$\pm$1.66 GHz. Given the value of s_2^-, we can extract a wavelength of 132.28$\pm$32 nm

301 which corresponds to the periodicity of where new domain wall appears into the system induced

15

302 by the wake of the domain-wall motion.

303   Interestingly, we also found that in principle there is no spatial limit for the propagation of

304 the shock waves in the systems. Simulations undertaken at a mesoscopic level (Supplementary

305 Figure 4) showed that once an electrical excitations is applied into the system, the transmission

306 of short-wavelength spin waves can extend for long-distances with minor disturbance from the

307 medium. We record lengths up to 10 μm with a group velocity above $\sim$2000 m s^{-1} where spin-

308 waves can work as nanoscale information carriers. The spatial distance is at the same order of

309 magnitude as those previously obtained in the magnetic insulator yttrium iron garnet (YIG)[68], and

310 ferromagnetic nanowires grown on thick YIG thin-film[69], with both have been proposed as plat-

311 forms to controllable transmission of spin information. The combination between small damping

312 ($\approx 2 \times 10^{-3}$) in this class of 2D ferromagnets[37], the insulating nature of CrI_3 and $CrBr_3$ with no

313 conduction electrons available to induce decay in the spin signal[70] and the relative low magnon

314 scattering[71] make magnetic layered materials a new endeavour for magnonics applications.

315 **Discussion**

316 The possibility to manipulate and control domain walls precisely on 2D vdW CrX_3 (X=Br, I)

317 magnets opens up a pathway to design a range of novel and highly competitive applications in

318 the thickness of a few atoms. As some of the critical parameters to domain-wall based devices

319 are domain wall widths, which control the information density, domain wall motion, directly gov-

320 erning access time, and pinning processes, that determine the energy consumption, 2D magnets

16

321 show features that are fundamental for integration in novel memory technologies, such as race-

322 track memories[4]. The large domain wall speeds achieved in both $CrBr_3$ and CrI_3 at the limit of

323 1020 m s^{-1} compete side-by-side with the best materials used for racetrack applications (e.g. syn-

324 thetic antiferromagnets[12]) where complex interfaces need to be fabricated to induce fast domain

325 wall shifting. This remarkable result, along with recent advances in the scalable bottom-up growth

326 of CrX_3 in the monolayer regime[72,73], clearly presents as an opportunity for reduction on man-

327 ufacturing and time-consuming device preparation. There is a still a challenge that needs to be

328 overcome to enable current-driven excitations in these 2D magnetic halides, for instance, the en-

329 hancement of the conductivity and the consequent reduction of the current densities used to move

330 domain walls. One route is to use doping, which is a well-known strategy that has been widely

331 employed in other prototypical ferromagnetic semiconductors (such as (Ga,Mn)As)[47]. In fact,

332 carrier doping has been found to induce half-metallicity and enhance the ferromagnetic stability

333 in CrI_3 monolayer[39,74], such that current-induced phenomena become feasible without compro-

334 mising the magnetic properties. It is worth mentioning that some trade-off between conductivity

335 enhancement and dissipation through the conduction electrons would have to be achieved for low-

336 power consumption. It is well known that large Gilbert damping λ_G allows fast relaxation of the

337 magnetization to equilibrium, whereas low damping enables energy-efficient processes. Several

338 approaches[75–78] were previously developed on metallic systems that may assist in the pursue of the

339 modelling of conduction electrons on doped-2D magnets. If the electrical conductivity of CrX_3

340 (X=I, Br) is enhanced due to different chemical or physical processes, the values of the domain

341 wall speeds shown in Figure 3 would be robust against such variations. Since the magnitude of

17

342   the Gilbert damping λ_G utilised in the simulations ($\lambda_G = 0.10$) is high than that expected for CrI_3

343   and $CrBr_3$[37], we intrinsically take into account additional dissipation effects in our simulations.

344   Furthermore, investigations to clarify the effect of different dopants, concentration and their im-

345   plications on both damping and the magnetic properties of 2D magnets are promptly needed. The

346   emission of spin waves in the THz regime and the spin hydrodynamic behaviour observed for both

347   halides, also merit further exploration on novel information technologies. On one hand, the de-

348   sign of sources, detectors and prototypes functioning in the THz-gap ($0.3 - 30$ THz)[79] provides the

349   groundwork for the developments of high-end electronics on the low-end of the electromagnetic

350   spectrum. The ultrathin character of 2D magnets, their flexibility in generating tunable frequen-

351   cies, and inexpensive sample cost relative to expensive bulky compounds[79], make vdW layered

352   materials a playground for applications. On the other hand, the use of magnons for information

353   carrier at the μm-scale puts vdW sheets in the roadmap of post-semiconductor spin-wave tech-

354   nologies. The characteristic soliton features observed on the shock-waves in both ferromagnets

355   and consequent low dissipation indicate that magnon-based data operations may be explored for

356   information processing. In this sense, the development of an energy-efficient spin-wave transducer

357   through vdW materials is one of the key steps for the ultimate goal of hybrid spin-wave computing

358   systems.

359   **Supplementary Materials**

360   Methods, Supplementary Figures S1−S4, and Supplementary movies S1−S12.

18

Data Availability

The data that support the findings of this study are available within the paper and its Supplementary

Information.

Competing interests

The Authors declare no conflict of interests.

Acknowledgments

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598 **Figure captions**

Figure 1: **Magnetic domains at different temperatures and fields. a-e,** Dynamical spin configurations of monolayer CrBr$_3$ during zero-field cooling (0 mT) at different temperatures. The out-of-plane magnetization M$_z$ (a.u.) is used to follow the evolution of the magnetic domains from 40 K down to 0 K. **f-j, k-o,** and **p-t,** Similar as **a-e,** but at magnetic fields of 5 mT, 11 mT and 50 mT, respectively. Temperatures at the top row correspond to all panels in the same column. Time scale spanned up to 2.5 ns till 0 K is achieved. Further evolution are observed at later times as shown in Supplementary Movies S1-S4.

Figure 2: **Hybrid domain walls with Néel-Bloch characteristics. a-b,** Global and local views, respectively, of a snapshot for one of the spin configurations projected along M$_z$ (colour map) showing the formation of a domain wall in monolayer CrBr$_3$. The total strip in **a** is 400×50 nm. Only Cr atoms are showed in the honeycomb lattice arrangement of monolayer CrBr$_3$. The system is at 0 K without any external fields or currents. **c-d,** Side and top views, respectively, of the rotation of the magnetization along the domain wall. Both M$_x$ and M$_y$ show variations along the wall altogether with M$_z$ which indicate a hybrid character of the domain wall with Néel and Bloch features. Colours follow the scale bar in **a**. The small area around the domain wall highlighted in **a** corresponds to **c**. **e-f,** Variation of the magnetization along the domain wall projected along the in-plane components (M$_x$, M$_y$) and M$_z$, respectively. Fitting lines are obtained using Eqs.2-3.

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Figure 3: **Current- and field-induced domain wall dynamics in 2D ferromagnets. a,** Initial domain wall configuration for monolayer CrBr$_3$ at no external driving forces (j=0, B=0). The domain wall is initially equilibrated for up to 4 ns to ensure convergence of the spin orientations along the wall and width. The same final configuration is utilised for both field- and current-driven domain wall motion. Similar procedure is undertaken to monolayer CrI$_3$. **b-c,** Snapshots of the domain wall dynamics in CrBr$_3$ induced by magnetic fields (0.05 T, 1.50 T) and electric currents (8.7 $\times$ 10^9 A cm^{-2}, 4.3 $\times$ 10^9 A cm^{-2}), respectively. The edges used in the simulations are highlighted at B=1.50 T with dangling-bond and zig-zag at the top and bottom of the layer, respectively. Deformations observed near the domain wall at j =4.3 $\times$ 10^9 A cm^{-2} are highlighted by the dashed rectangle. **d,** Zoom-in of the local spin-configurations at j =4.3 $\times$ 10^9 A cm^{-2}) with colour scale showing the variations of the spin orientations along M$_z$ induced by the large current density.

Figure 4: **Domain wall speeds and spin wave frequency. a-b,** Calculated domain wall velocities v versus the applied magnetic field B for CrBr$_3$ and CrI$_3$, respectively. The vertical solid lines indicate the region where the nucleation of magnetic bubbles starts in each material as those shown in **b** at 1.50 T for CrBr$_3$. A fit using $v = \mu(\text{B} - \text{B}_\text{o})$, where μ is the domain wall mobility and B$_\text{o}$ is the onset field, reproduces accurately the atomistic simulation data with linear regression coefficients R^2 = 0.99 and a root-mean-square-error of 0.0074. The field is applied perpendicular to the surface following the easy-axis of the materials. **c-d,** Variations of v as a function of the current density j for CrBr$_3$ and CrI$_3$, respectively. Two regimes are observed in the internal dynamics of the wall before and after the Walker breakdown (j_w) with the wall motion being in steady and precessional states, respectively. We could extract $j_w^{CrBr_3}$ =4.8 $\times$ 10^9 A cm^{-2} and $j_w^{CrI_3}$ =12.0 $\times$ 10^9 A cm^{-2}. Both steady and precessional regimes on the domain-wall dynamics can be modelled using a 1D model as described in the text. The insets show the velocities at lower values of current within $j \leq 8.5 \times 10^8$ A cm^{-2}. **e-f,** Frequencies ω_B(THz) and ω_J(THz) of the emitted spin-waves during the domain-wall motion as a function of B and j, respectively, for CrBr$_3$ and CrI$_3$. Dashed lines are corresponding fits to Eqs. 5−6. Similar data labelling applies for **e** and **f**.

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Figure 5: **Modulational instability in 2D magnets.** **a-c,** Snapshots of the magnetisation along M_z and M_x magnetisation components at different times at 18 ps, 40 ps and 78 ps, respectively. **d,** Amplitude of M_x as a function of time at different current densities.

Figure 6: **Long range spin-wave propagation. a-c,** Snapshots of spin-dynamics at 20 ps, 200 ps and 600 ps, respectively, under a current density of 1.56×10^{10} A cm^{-2}. M_z is used to show the variations of the domain wall with the colour scale along the length of the ribbon. A separation of the motion of the original domain wall (soliton edge) and the shock waves can be observed between a wave train (modulationally unstable) propagating throughout the system. **d,** Colour-plot of the variations of M_z with the propagation time along a 2-μm ribbon of CrBr$_3$ at a current density of 1.56×10^{10} A cm^{-2}. The different line-cuts correspond to the several waveforms moving into the systems as shown in **e-g**. The dashed lines are given by $s^+ = 5773$ m s^{-1}± 0.84 m s^{-1}, $s_1^- = 1328$ m s^{-1}± 1.7 m s^{-1} and $s_2^- = 882$ m s^{-1}± 1.8 m s^{-1}. **e-f,** Spectral density (arb. units) extracted from the fast Fourier transform of the M_z component as function of time in **d** along velocities s^+ and s_2^-, respectively. The characteristic frequencies of the shock waves and the domain-wall wake can be identified as $\nu_{SW} = 177.5$ GHz± 1.25 GHz and $\nu_{wake} = 6.66 \pm 1.66$ GHz, respectively.

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599 **Figures**

Figure 1

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Figure 2

Figure 3

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Figure 4

Figure 5

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Figure 6