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### Domain wall dynamics in two-dimensional van der Waals ferromagnets

- Dina Abdul-Wahab<sup>1</sup>, Ezio Iacocca<sup>2,3</sup>, Richard F. L. Evans<sup>4</sup>, Amilcar Bedoya-Pinto<sup>5</sup>, Stuart Parkin<sup>5</sup>,
- Kostya S. Novoselov<sup>6,7</sup> & Elton J. G. Santos<sup>8,9†</sup>
- <sup>1</sup>School of Mathematics and Physics, Queen's University Belfast, BT7 1NN, UK
- Center for Magnetism and Magnetic Materials, University of Colorado at Colorado Springs, Col-
- orado Springs, CO 80918, USA
- <sup>3</sup>Department of Mathematics, Physics, and Electrical Engineering, Northumbria University, New-
- castle upon Tyne, NE1 8ST, UK
- <sup>4</sup>Department of Physics, The University of York, YO10 5DD, UK
- <sup>5</sup>NISE Department, Max Planck Institute of Microstructure Physics, Halle, Germany 11
- <sup>6</sup>Department of Material Science & Engineering, National University of Singapore, Block EA, 9 12
- Engineering Drive 1, 117575, Singapore 13
- <sup>7</sup>Chongqing 2D Materials Institute, Liangjiang New Area, Chongqing 400714, China
- Institute for Condensed Matter Physics and Complex Systems, School of Physics and Astronomy,
- The University of Edinburgh, EH9 3FD, UK.
- Higgs Centre for Theoretical Physics, The University of Edinburgh, EH9 3FD, UK
- Corresponding author: esantos@ed.ac.uk
- Domain wall motion is in the core of many information technologies ranging from storage<sup>1</sup>,
- processing<sup>2</sup>, sensing<sup>3</sup> up to novel racetrack memory architectures<sup>4</sup>. The finding of magnetism

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in two-dimensional (2D) van der Waals (vdW) materials<sup>5-8</sup> has offered a new frontier for the exploration and understanding of domain walls at the limit of few atom-thick layers. Howver, to use 2D vdW magnets for building spintronics nanodevices such as domain-wall based 23 logic<sup>9-11</sup>, it is required to gain control of their domain wall dynamics by external driving 24 forces such as spin-polarized currents or magnetic fields which has so far been elusive. Here 25 we show that electric currents as well as magnetic fields can efficiently move domain walls in the recently discovered 2D vdW magnets CrI<sub>3</sub> and CrBr<sub>3</sub> at low temperatures, and robust 27 down to monolayer. We realize field- and current-driven domain wall motion with velocities up to  $1020 \text{ m s}^{-1}$  which are comparable to the state-of-the-art materials for domain-wall 29 based applications 12-16. Domain walls keep their coherence driven by the spin-transfer torque 30 induced by the current and magnetic field up to large values of about  $12 \times 10^9$  A cm<sup>-2</sup> and T, respectively. For larger magnitudes of current or field, a transition to a hydrodynamic spin-liquid regime is observed with the emission of a periodic train of spin-wave solitons 33 vith modulational instability<sup>17</sup>. The emitted waveform achieves terahertz (THz) frequency 34 in a wide range of field and current densities which opens up perspectives for reconfigurable 35 nagnonic devices. Moreover, we found that these spin-waves can transport spin angular momentum through the layers over distances as long as 10 μm without losses for the transport of spin information. Our results push the boundary of what is currently known about the 38 dynamics of domain walls in 2D vdW ferromagnets and unveil strategies to design ultrathin, high-speed and high-frequency spintronic devices.



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storage technologies<sup>18,19</sup>. A promising strategy is to encode bits in magnetic domains walls which an be controlled via applied magnetic fields and spin-polarized currents<sup>4,9</sup>. Several approaches or domain wall-based memory devices with a focus on the domain wall displacement have been developed. From a field-controlled shift register<sup>9,20</sup> up to the non-volatile multi-turn sensors<sup>21</sup>, which are commercially available<sup>22</sup>, these emerging technologies provide a new horizon for ad-

Spin-based applications have been broadly explored for high-performance solid-state data

vanced materials to be explored. With the discovery of magnetism in vdW layered compounds<sup>5–7</sup>

and proof-of-concept devices<sup>8,23-25</sup> 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<sup>26</sup>, competitive devices should have 50 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 52 100 m s<sup>-1</sup> to achieve rapid operating speed on increasing areal density<sup>4,27</sup>. Sufficiently low 53 urrent densities ( $\sim 10^6 - 10^8 \text{ A cm}^{-2}$ ) are also needed to guarantee low power consumption and avoid damage due to Joule heating. However, metallic layers inherently suffer with energy dissipation via the conduction electrons<sup>2</sup> which make magnetic insulators better suited for domain all applications. There have been a few reports on the creation of different interfaces using magnetic vdW materials<sup>28,29</sup> 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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would be in domain-wall based functional devices.

Here we demonstrate domain-wall motion in monolayer CrI<sub>3</sub> and CrBr<sub>3</sub> by spin polarized 63 currents and magnetic fields in a broad range of densities  $(10^7 - 10^9 \text{ A cm}^{-2})$  and field (0.001 - 2)T). Both dynamics occur over hybrid domain walls with Néel-Bloch characteristics with a domain 65 wall width of about ~5.30 nm. The spin-torque induced by electric currents and magnetic fields generates a rapid rotation of the magnetization at the domain wall, hence triggering its translational motion at high velocities. We record speeds of up 1020 m s<sup>-1</sup> and 100 m s<sup>-1</sup> with current and field, respectively, while keeping the coherence of the wall profile through a steady motion. As the domain wall velocity approaches the maximum spin-wave group velocity, the domain wall motion start to exhibit non-linear effects with the emission of magnons at terahertz (THz) frequencies following a modulational instability behaviour. Even though no conduction electrons are resent in the medium due to the insulating characteristics of CrI<sub>3</sub> and CrBr<sub>3</sub>, spin-information can be transported over distances as long as 10 μm which sets a new paradigm for spin-wave-based 74 technologies at the ultrathin limit.

Our starting point is the investigation of the magnetic domain structures in monolayer CrBr<sub>3</sub> and their dynamic evolution under different temperatures and magnetic fields. The domain wall structure of CrI<sub>3</sub> and its cooling dynamics have recently been reported using similar theoretical 78 framework<sup>30</sup>. We use a large square flake of 0.4  $\mu$ m imes 0.4  $\mu$ m to represent a system able to be measured using imagining techniques, i.e. nitrogen-vacancy scanning magnetometry<sup>31</sup>. We describe the interactions using the following spin Hamiltonian:

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

where  $S_i$  and  $S_j$  are the localized magnetic moments on Cr atomic sites i and j which are coupled y pair-wise exchange interactions.  $J_{ij}$  and  $\lambda_{ij}$  are the isotropic and anisotropic bilinear (BL) exhanges, respectively, and  $D_i$  is the single ion magnetic anisotropy.  $\mathbf{B}_i$  and  $\mathbf{B}_i^{\mathrm{dp}}$  represent external and dipole magnetic field sources respectively. Eq. 1 was previously found to describe accurately the magnetic properties of several 2D magnets<sup>30,32</sup> including CrI<sub>3</sub> and CrBr<sub>3</sub>. Comparisons with other spin Hamiltonians such as Kitaev, bilinear Heisenberg, Ising, and Dzyloshinskii-Moriya interactions, were undertaken in refs. 30,32 as well as in their Supplementary Information files. We take into account up to third nearest neighbors for  $J_{ij}$  and  $\lambda_{ij}$  in the description of CrBr<sub>3</sub> in Eq.1. All the parameters in Eq.1 are extracted using highly accurate non-collinear ab initio simulations taking into account spin-orbit coupling and Hubbard-U corrected density functional theory as described in Ref.  $^{30,32}$ . We ensure that fine numerical convergence within  $10^{-6} - 10^{-8}$  eV is achieved in each computed parameter. We also compared the exchange parameters used in our work with those collected from the literature<sup>33</sup>. In the context of the materials studied in our study (monolayer CrI<sub>3</sub> and CrBr<sub>3</sub>), Supplementary Figure 1 clearly shows that our exchange magnitudes<sup>32</sup> reproduce with high accuracy the critical temperatures measured. Moreover, Eq.1 provides an accurate decription of the magnon dispersion extracted from inelastic neutron scattering measurements on CrI<sub>3</sub><sup>34</sup> as shown in ref.<sup>32</sup>. These results provide a firm background for the modeling of the domain wall dynamics as described here.

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

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of two or more electrons between two adjacent sites<sup>32</sup>. Its strength is given by the constant  $K_{ij}$ , 101 which is the simplest and most natural form of non-Heisenberg coupling. It has recently been found that several 2D vdW magnets develop substantial BQ exchange in their magnetic properties which 103 critical to quantitatively describe important features such as Curie temperatures, thermal stability, 104 magnon spectra and non-trivial topological spin textures  $^{30,32,35,36}$ . The magnitude of  $K_{ij}$  for CrBr<sub>3</sub> 105 is 0.22 meV which is smaller than the BL exchange for the first-nearest neighbours ( $J_1 = 1.66 \text{ meV}$ ) but still sizeable to be disregarded<sup>32</sup>. In our implementation the BQ exchange is quite general and 107 an be applied to any pair-wise exchange interaction of arbitrary range. Each system is evolved by 108 solving the atomistic Landau-Lifshitz-Gilbert equation with a Gilbert damping  $\lambda_G=0.1$  to ensure 109 better computational efficiency and numerical stability of the time integration. The value of  $\lambda_G$  is 110 substantially higher than that  $(2 \times 10^{-3})$  recently measured for a parent compound<sup>37</sup> (i.e. CrCl<sub>3</sub>) 111 which a similar magnitude would be expected for  $CrI_3$  and  $CrBr_3$ . The large  $\lambda_G$  also accounts 112 or the possibility of free-electron doping to raise the conductivity of the system due to different 113 rocesses (i.e. dopants<sup>38</sup>, electric bias<sup>39</sup>). Due to the inclusion of dipole-dipole interactions we 114 explicitly include non-local damping effects in our simulations such as due to domain walls or 115 different magnetic textures. Hence, our spin-dynamics simulations include an accurate description of the spin-interactions at the atomistic level (2-20 Å) in a multiscale framework at the lab scale (a few µm's). 118 Monolayer CrBr<sub>3</sub> is thermally equilibrated above 40 K and then linearly cooled to 0 K in 119

a total simulated time of 4.0 ns at different magnetic fields (Figure 1 and Supplementary Movies 120 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 122 (0 mT) magnetic domains are formed for temperatures below 20 K with the free motion of the domain walls as the temperature drops (Fig. 1a-e). As the system reaches 0 K a continuous 124 time evolution of the domain structures remains which results in a homogenous magnetization 125 over the entire crystal (Supplementary Movie S1). Even though thermal fluctuations as well as 126 their contributions to the energy of the system are inexistent at 0 K, other terms, such as dipolar 127 fields, exchange interactions and magnetic anisotropy, are still present, further contributing to the 128 modification of the magnetic domains. Once magnetic fields are applied to the nanosheet (Fig. 129 1f-t and Supplementary Movies S2-S4) this nucleation-type mechanism is accelerated with a rapid 130 reversal in the sign of the magnetization M<sub>z</sub>. Indeed, a monodomain feature is observed even 131 in fields far below the coercivity<sup>40</sup> (10 mT at 5 K) for CrBr<sub>3</sub>. In a scenario where defects or 132 pinning sites are not present in monolayer CrBr<sub>3</sub>, magnetic domains tend to be meta-stable with 133 homogeneous magnetisation throughout the surface. This is in sound agreement with recent 134 V-center scanning magnetometry measurements<sup>31</sup> on the magnetic domain evolution of CrBr<sub>3</sub> 135 indicating that pinning effects are the dominant coercivity mechanisms.

Strikingly, the interplay between domain metastability and high magnetic anisotropy of 137 CrBr<sub>3</sub> give additional characteristics to the domain walls (Figure 2a-b). We observe that as the 138 spins vary orientation from one magnetic domain to another the wall profile assumes components 139 of the magnetization along different in-plane directions  $(M_{x,y})$  relative to the easy-axis  $(M_z)$  (Fig. 140 **2c-d**). In-plane magnetization  $M_x$  displays a variation larger than that for  $M_y$  which is mainly no-141 ticed at the core of the domain wall within 1-2.5 nm. These features indicate a domain wall with

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hybrid characteristics of Bloch and Néel type (Fig. 2e-f). We can extract the domain wall width  $\sigma_{x,y,z}$  by fitting the different components of the magnetization  $(M_x,M_y,M_z)$  to standard equations of the form<sup>41</sup>:  $M_j = \frac{1}{\cosh(\pi(j-j_0)/\sigma_j)}$ , with j = x, y(2)  $M_z = \tanh(\pi(z-z_0)/\sigma_z)$ (3)

where  $j_0$  and  $z_0$  are the domain wall positions at in-plane and out-of-plane coordinates, respec-146 tively. The domain wall widths are within the range of  $\sigma_{x,y,z} = 5.30 - 5.33$  nm. Materials with 147 high magnetic anisotropy generally stabilise such small domain wall widths but their magnetic domains are generally stable after zero-field cooling due to long range dipole interactions. We have 149 checked that the inclusion of dipolar fields in our simulations do not change the results. 150

An intriguing question raised by the hybrid features of the domain walls in CrBr<sub>3</sub> and CrI<sub>3</sub><sup>30</sup> 151 is the effect of magnetic fields and electric currents on the motion of domain walls. It is well estab-152 lished that both driving forces can induce displacement of domain walls in magnetic thin films over 153 different substrates. Nevertheless, the vdW nature of the nanosheet together with the atomic thick-154 ness ( $\approx$ 0.5 nm) may induce additional phenomena yet to be observed in 2D ferromagnets. Figure 155 3a-d shows that either fields or currents can efficiently displace domain walls in layered CrBr3 and 156 CrI<sub>3</sub> magnets. The dynamics is generally initialised with a domain wall at equilibrium position 157 previously thermalized for several nanoseconds for full convergence of the spin orientations (Fig. 3a). The finite width of the nanowire assists in the formation of the domain walls pinned by the 159 edges. For field-induced domain wall motion, wall velocities up to  $v = 70 \text{ m s}^{-1}$  and  $v = 98 \text{ m s}^{-1}$ 



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are recorded for CrBr3 and CrI3 (Fig. 4a-b), respectively. The wall moves steadily on both sheets and can be described as  $v = \mu_F(B - B_0)$ , where  $\mu_F$  is the wall mobility and  $B_0$  is the onset field required to move the wall from structural defects or pinning<sup>42</sup>. The magnitudes of  $\mu = 0.0343$  m 163  $^{-1}$  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 $_{3}$ 164 indicate that domain walls move relatively slower than those in metallic ultrathin Pt/Co/Pt films<sup>43</sup> 165 or thick magnetic insulator TmIG/Pt interfaces<sup>14</sup> but with relatively similar onset field B<sub>0</sub>. The domain wall moves steadily on both sheets at fields below critical magnitudes ( $B_c$ ) of  $B_c = 1.19$ 167 and B<sub>c</sub> = 4.78 T for CrBr<sub>3</sub> and CrI<sub>3</sub>, respectively (Supplementary Movies S5-S6). For values 168 beyond B<sub>c</sub> different spin distributions start nucleating ahead of the domain-wall (Supplementary 169 Movie S7-S8) and induce its collapse within a few hundreds of picoseconds after the motion ini-170 tiated. These spin features appear from the edges of the layers and rapidly move into the bulk of 171 the ribbon. This behaviour is analogous to that observed in Pt/Co/AlO<sub>x</sub> ultrathin microstructures<sup>44</sup> 172 and highlighted the importance of edges on the magnetic properties of 2D magnets in device in-173 tegration. A preference for unreconstructed edges or dangling bonds rather than on zig-zag edges 174 (Fig. 3b at 1.50 T) has been observed in the nucleation of reversed domains during the wall dy-175 namics. The local variation of the exchange interactions at dangling-bonds with spins being less restricted to change their directions with an external field than at zig-zag edges is one of the main 177 ingredients for this behaviour. This observation shall trigger further experimental studies of the 178 atomic structure at the edges of the monolayer magnets via scanning probe microscopies<sup>45,46</sup> and 179 their connection to the device performance.

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



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complished in two ways. First, by using an adjacent conducting layer as recently demonstrated for 182 insulating Tm<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> on top of a Pt substrate<sup>14</sup>. Charge current will flow in the heavy metal generating spin currents that exert a spin-transfer torque (STT) on the ferromagnetic material. Second, 184 by doping the CrX<sub>3</sub> (X=Br, I) magnets in order to generate significant carriers in the host<sup>27,47</sup>. Such 185 strategies are well established and would provide a feasible platform to confirm our predictions. 186 Indeed, we recorded domain wall velocities up to 530 m s<sup>-1</sup> and 1020 m s<sup>-1</sup> for monolayer CrBr<sub>3</sub> 187 and CrI<sub>3</sub>, respectively, under applied currents (Fig. 4c-d). These velocities are higher than those 188 observed in a wide range of systems including skyrmions (100 m s<sup>-1</sup>)<sup>13</sup>, ferromagnetic semicon-189 ductor (Ga, Mn)As (22 m s<sup>-1</sup>)<sup>47</sup>, synthetic antiferromagnets (750 m s<sup>-1</sup>)<sup>12</sup>, metallic layers (380 m 190  $^{-1}$ )<sup>48</sup>, Pt/CoFe/MgO and Co/Ni/Co interfaces ( $\sim$ 10 m s<sup>-1</sup>, 400 m s<sup>-1</sup>)<sup>16,49</sup>, and insulating oxides 191 (400 m s<sup>-1</sup>)<sup>14</sup>. It is worthwhile highlighting that low current densities within the experimental 192 range of  $10^{-8}$  A cm<sup>-2</sup> (insets in Fig. 4**c-d**) already resulted in domain wall speeds (i.e.  $100 \text{ m s}^{-1}$ ) similarly as those achieved in established racetrack platforms with more complex materials<sup>4</sup>. The 194 domain walls displace freely as a function of the current density j in a viscous flow motion (Supple-195 mentary Movies S9-S10). The high velocities predicted for CrBr<sub>3</sub> and CrI<sub>3</sub> are only limited by the spin-wave group velocity  $v_g$  intrinsic to the systems. By using a spin-wave theory for 2D magnetic materials<sup>32</sup>, which includes biquadratic exchange and Dzyaloshinskii-Moriya interactions (DMI), we estimate from the magnon dispersion (Supplementary Figure 2) maximum values of  $v_g$  within 199 the range of 584–891 m s<sup>-1</sup> for CrBr<sub>3</sub> and 1326–6400 m s<sup>-1</sup> for CrI<sub>3</sub> (Supplementary Figure 3). These values are close to those computed from the atomistic simulations (530 m s<sup>-1</sup> and 1020 m s<sup>-1</sup> for CrBr<sub>3</sub> and CrI<sub>3</sub>, respectively) although slightly larger. This is due to the damping of

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the domain wall motion by the emission of spin-waves<sup>50</sup> not taken into account into the model as commented in the following.

The steady displacement of the domain walls occurs up to a certain threshold  $(j_w)$  defined as the Walker breakdown<sup>42</sup>. For values below  $j_w$ , the wall dynamics can be well modelled by a simple 1D model<sup>51,52</sup> through a linear dependence of v and j:

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

where  $\mu_I$  is the current-driven domain-wall mobility which gives  $\mu_I = 0.12 \times 10^{-10} \text{ m}^3 \text{ A}^{-1} \text{s}^{-1}$ and  $\mu_I = 0.10 \times 10^{-10} \text{ m}^3 \text{ A}^{-1} \text{s}^{-1}$  for monolayer CrBr<sub>3</sub> and CrI<sub>3</sub>, respectively. These values 206 are of the same order of magnitude as those recorded on thin metallic interfaces<sup>49</sup> or permalloy 207 nanowires<sup>53</sup> which have been studied more intensively. For values near  $j_w$  or above, the spins 208 at the domain wall precesses inducing disruption of the wall at longer times with the nucleation of magnetic domains with a different polarisation ahead of the displacement of the domain-wall (Fig. 3c at  $4.35 \times 10^9$  A cm<sup>-2</sup>). A close look at these features (Fig. 3d) indicates that they 211 may start from both edges although dangling-bonds may induce more curvature to the domain 212 wall since a dragging on the motion is observed generating retardation relative to the zig-zag edge. 213 We can model qualitatively the dynamics at  $j > j_w$  for both CrBr<sub>3</sub> and CrI<sub>3</sub> via the time average velocity of the domain wall<sup>51,52</sup>. This suggests that the domain-wall dynamics in 2D magnets is 215 similar as that in magnetic nanowires which allows faster integration into existing devices<sup>27</sup>. We 216 also observe that as the domain wall moves spin-waves or magnons are emitted (Fig. 4e-f). The 217 spin frequencies driven by the field  $(\omega_R)$  and current  $(\omega_I)$  are in the terahertz (THz) regime with 218 maximum magnitudes of  $\omega_B^{max}=0.43$  THz and  $\omega_J^{max}=0.60$  THz for CrBr3 and  $\omega_B^{max}=0.50$  THz

and  $\omega_J^{max} = 0.66$  THz for CrI<sub>3</sub>. These frequencies are in sound agreement with those estimate from the magnon dispersion along different points of the Brillouin zone for both halides (Supplementary Figure 3). We found that the maximum frequencies with the field are close to those at the M-point (0.47 THz for CrBr<sub>3</sub>, and 0.57 THz for CrI<sub>3</sub>), whereas with the applied current are around the K-point (0.63 THz for CrBr<sub>3</sub>, and 0.64 THz for CrI<sub>3</sub>). Indeed, the variations of  $\omega_B$  and  $\omega_J$  can be well fitted using:

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

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

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  $\omega_0^J=0.31$  THz for CrBr<sub>3</sub>; and  $\omega_0^B=0.25$  THz and  $\omega_0^J=0.43$  THz for CrI<sub>3</sub>. We also notice 227 that the magnitudes of  $\omega_0^B$  and  $\omega_0^J$  may also change due to the inhomogeneous spatial distribution 228 of the emitted spin-waves (Supplementary Movie 11) even though with a similar behaviour with 229 the field and current. The coefficient  $\alpha_B$  differs moderately between both halides converging to 0.13 THz T<sup>-1</sup> and 0.05 THz T<sup>-1</sup> for CrBr<sub>3</sub> and CrI<sub>3</sub>, respectively, with  $\alpha_J \approx 0.04$  THz  $10^{-9}$  A 231 cm<sup>-2</sup> being similar to both systems. On the field-driven spin-wave emission, the domain wall pre-232 cesses with a frequency near the Larmor frequency  $\omega_0 = \gamma B$ , where  $\gamma$  is the gyromagnetic ratio, 233 similarly as in nanowires<sup>54</sup>. We noticed that in both layered materials the spin wave frequencies 234 are enclosed between odd- and even-numbered harmonics. That is, CrBr<sub>3</sub> is within  $4\omega_0$  and  $5\omega_0$  $(\omega_B - \omega_o^B = 4.81\omega_0)$ , and CrI<sub>3</sub> is within  $\omega_0$  and  $2\omega_0$   $(\omega_B - \omega_o^B = 1.70\omega_0)$ . This indicates that the 236 spin waves and their overtones can be excited with a combination of fundamental modes which 237 gives additional flexibility for terahertz source of magnetic signal using atomically thin layers. In

addition, the emission of spin waves electrically-driven can be associated with the Doppler effect on the frequency shift<sup>55</sup>. As the domain wall moves the current changes the spin-waves dispersion and incidentally causes an effective flow of the magnetic medium. This is reflected by the linear dependence of  $\omega_J$  on j (Fig. 4f) in agreement with the spin-transfer-torque Doppler shift. A similar approach has been successfully used to study the effect of electric signal into magnons in permalloy strips<sup>56</sup>, Ni<sub>80</sub>Fe<sub>20</sub> wires<sup>57</sup> and thin films<sup>58</sup>.

A substantially different phenomenon is observed when the domain wall is subject to large 245 current densities,  $j > 10^{10} \text{ A cm}^{-2}$ , with the appearance of spin hydrodynamic effects in CrBr<sub>3</sub> 246 and CrI<sub>3</sub>. The dynamics is illustrated in Figure 5a-c where we plot line-cuts of the M<sub>z</sub> and M<sub>x</sub> 247 components of the magnetisation at width 25 nm and selected times for monolayer CrI<sub>3</sub>. We 248 observe that shortly after the electric pulse starting propagating into the system within 18-78 ps the M<sub>x</sub> projection exhibits a wave propagating ahead of the domain wall. This amplitude tilts 250  $M_z$  into the plane and increases exponentially (Fig. 5d) until  $M_z$  is completely suppressed and 251 eventually switches, nucleating a new domain wall. The first period of the wave in  $M_x$  or shock 252 wave continues to propagate with no appreciable dissipation and new domain walls are nucleated 253 its wake with a certain periodicity (Fig. 5a-c). This nonlinear phenomenon is indicative of a modulational instability (MI) as observed in fluid dynamics<sup>17,59</sup>. This type of instability occurs in focusing media and describes the exponential growth of waves within a band of wavevectors until 256 non-linear effects favour their spatial localisation into solitons. MI are ubiquitous in dispersive 257 physical problems and have been observed in several systems such as in water waves<sup>60</sup>, nonlinear optics 61 and Bose-Einstein condensates 62. In magnetic materials however MI has been invoked

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so far as a pathway for the nucleation of dissipative droplets in perpendicular magnetic anisotropy (PMA) ferromagnets<sup>63</sup>, as well as the spatial localisation of high-energy magnons in PMA ferrimagnetic alloys excited with ultrafast optical pulses<sup>64</sup>, and the disintegration of short-wavelength 262 spin-superfluids in planar ferromagnets<sup>65</sup>. In contrast, we find clear evidence of spin hydrodynamic 263 behaviour manifested the onset of MI and its nonlinear dynamics driven by electrical currents in 264 2D magnetic materials. We emphasize that the nonlinear effect observed here is fundamentally different from Walker breakdown and the subsequent precessional regime so far observed in bulk magnets. 267 We also observe that MI is visible already at 20 ps as the wavefront exhibits a transverse

268 structure and can be more generally regarded as a spin shock wave (Fig.6a-c). At 200 ps, there are rich transverse features in the nanoribbon, from which we can identify a modulationally unstable region where new domains are established. These domains are bounded by the trailing domain-wall 271 soliton edge) and the spin shock wave. At 600 ps, the shock wave is already outside our simulation 272 rofile and the domain walls continue to propagate further (see Supplementary Movie S12 for the 273 full sequence). The salient features of the MI dynamics can be visualized in Fig. 6d by a colour-plot 274 of the M<sub>2</sub> component line-cuts as a function of time. After an initial translation of the domain wall, there is a split of behaviours at  $\approx 0.01$  ns where we can define two characteristic speeds (dashed lines in Fig. 6d). The shock wave (SW) (Fig. 6b) translates with a speed of  $s^+ = 5.77$  km s<sup>-1</sup>  $\pm$ 277 0.84 km s<sup>-1</sup> which is in sound agreement with the maximum group velocity calculated for CrI<sub>3</sub> 278 (Supplementary Figure 3). The soliton edge exhibits a phase-shift and subsequent translation with a speed of  $s_1^-=1.33~{\rm km~s^{-1}\pm0.002~km~s^{-1}}$  (Fig. 6b). Consequently, the wavelength L of the



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nucleated domains can be extracted from the spectral function at a constant speed, such at  $s^+$ (Fig. 6e). The obtained frequency of the wavefront or SW is computed as  $v_{SW}$  =177.5 GHz±1.25 GHz. We can immediately extract L through a simple speed and frequency relationship via L =283  $s^+/v_{SW}=5.77~{\rm km~s^{-1}}/177.5~{\rm GHz}=32.50\pm0.23~{\rm nm}$  which is equivalent to a wave-vector of 284  $k_{SW}=2\pi/L=0.19~{
m nm}^{-1}\pm0.00003~{
m nm}^{-1}.$  Indeed, the shock-wave speed corresponds to the 285 group velocity of the most unstable magnon with a wave-vector given by  $k_{SW}$ .

The growing region bounded by the speeds  $s^+$  and  $s_1^-$  is reminiscent to the development of 287 dispersive shock waves (DSWs)17 which is located at the modulationally unstable region high-288 lighted on Fig.6b. DSWs are ordered structures that smoothly connect the wavevectors at the front 289 and soliton edges, and have been fully characterised for planar ferromagnets<sup>66</sup>. However, the MI our system induces periodic domain-wall nucleation defined by the most unstable wavevector. Therefore, we can regard the scenario of Figs.6b-c as an unstable generalization of a DSW. 292 he periodicity of the nucleated domains leads to initially similar domain-wall speeds (parallel 293 features in the wake of the wavefront) that are later perturbed by interactions, annihilation, and 294 nucleation events. These can be potentially related to turbulence, thermodynamic behaviour as a 295 soliton gas<sup>67</sup>, or rare events such as rogue waves<sup>60</sup>. At a later time ( $\approx 0.04$  ns), a second characteristic domain-wall speed  $s_2^- = 0.88 \text{ km s}^{-1} \pm 0.001 \text{ km s}^{-1}$  is observed. This second speed appears to be related to an additional source of MI in the wake of the soliton-edge that opposes the 298 current-driven torque. We can extract the frequency of this domain-wall wake (Fig.6f) resulting 299 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 which corresponds to the periodicity of where new domain wall appears into the system induced

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by the wake of the domain-wall motion.

Interestingly, we also found that in principle there is no spatial limit for the propagation of 303 the shock waves in the systems. Simulations undertaken at a mesoscopic level (Supplementary 304 Figure 4) showed that once an electrical excitations is applied into the system, the transmission 305 of short-wavelength spin waves can extend for long-distances with minor disturbance from the medium. We record lengths up to 10  $\mu$ m with a group velocity above ~2000 m s<sup>-1</sup> where spinwaves can work as nanoscale information carriers. The spatial distance is at the same order of magnitude as those previously obtained in the magnetic insulator yttrium iron garnet (YIG)68, and ferromagnetic nanowires grown on thick YIG thin-film<sup>69</sup>, with both have been proposed as plat-310 forms to controllable transmission of spin information. The combination between small damping 311  $(\approx 2 \times 10^{-3})$  in this class of 2D ferromagnets<sup>37</sup>, the insulating nature of CrI<sub>3</sub> and CrBr<sub>3</sub> with no conduction electrons available to induce decay in the spin signal<sup>70</sup> and the relative low magnon 313 scattering<sup>71</sup> make magnetic layered materials a new endeavour for magnonics applications. 314

### Discussion 315

The possibility to manipulate and control domain walls precisely on 2D vdW CrX<sub>3</sub> (X=Br, I) magnets opens up a pathway to design a range of novel and highly competitive applications in the thickness of a few atoms. As some of the critical parameters to domain-wall based devices 318 are domain wall widths, which control the information density, domain wall motion, directly gov-319 erning access time, and pinning processes, that determine the energy consumption, 2D magnets

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show features that are fundamental for integration in novel memory technologies, such as race-321 track memories4. The large domain wall speeds achieved in both CrBr3 and CrI3 at the limit of 020 m s<sup>-1</sup> compete side-by-side with the best materials used for racetrack applications (e.g. syn-323 thetic antiferromagnets<sup>12</sup>) where complex interfaces need to be fabricated to induce fast domain 324 wall shifting. This remarkable result, along with recent advances in the scalable bottom-up growth 325 of CrX<sub>3</sub> in the monolayer regime<sup>72,73</sup>, clearly presents as an opportunity for reduction on manufacturing and time-consuming device preparation. There is a still a challenge that needs to be 327 overcome to enable current-driven excitations in these 2D magnetic halides, for instance, the en-328 hancement of the conductivity and the consequent reduction of the current densities used to move 329 domain walls. One route is to use doping, which is a well-known strategy that has been widely 330 employed in other prototypical ferromagnetic semiconductors (such as (Ga,Mn)As)<sup>47</sup>. In fact, 331 carrier doping has been found to induce half-metallicity and enhance the ferromagnetic stability 332 in CrI<sub>3</sub> monolayer<sup>39,74</sup>, such that current-induced phenomena become feasible without compro-333 mising the magnetic properties. It is worth mentioning that some trade-off between conductivity 334 enhancement and dissipation through the conduction electrons would have to be achieved for low-335 power consumption. It is well known that large Gilbert damping  $\lambda_G$  allows fast relaxation of the magnetization to equilibrium, whereas low damping enables energy-efficient processes. Several 337 approaches<sup>75–78</sup> were previously developed on metallic systems that may assist in the pursue of the 338 modelling of conduction electrons on doped-2D magnets. If the electrical conductivity of CrX<sub>3</sub> 339 (X=I, Br) is enhanced due to different chemical or physical processes, the values of the domain 340 wall speeds shown in Figure 3 would be robust against such variations. Since the magnitude of



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the Gilbert damping  $\lambda_G$  utilised in the simulations ( $\lambda_G = 0.10$ ) is high than that expected for CrI<sub>3</sub> and CrBr<sub>3</sub><sup>37</sup>, we intrinsically take into account additional dissipation effects in our simulations. Furthermore, investigations to clarify the effect of different dopants, concentration and their im-344 plications on both damping and the magnetic properties of 2D magnets are promptly needed. The 345 emission of spin waves in the THz regime and the spin hydrodynamic behaviour observed for both 346 halides, also merit further exploration on novel information technologies. On one hand, the de-347 sign of sources, detectors and prototypes functioning in the THz-gap  $(0.3-30 \text{ THz})^{79}$  provides the 348 groundwork for the developments of high-end electronics on the low-end of the electromagnetic 349 spectrum. The ultrathin character of 2D magnets, their flexibility in generating tunable frequen-350 cies, and inexpensive sample cost relative to expensive bulky compounds<sup>79</sup>, make vdW layered 351 materials a playground for applications. On the other hand, the use of magnons for information carrier at the µm-scale puts vdW sheets in the roadmap of post-semiconductor spin-wave tech-353 nologies. The characteristic soliton features observed on the shock-waves in both ferromagnets 354 and consequent low dissipation indicate that magnon-based data operations may be explored for 355 information processing. In this sense, the development of an energy-efficient spin-wave transducer through vdW materials is one of the key steps for the ultimate goal of hybrid spin-wave computing systems.

### **Supplementary Materials**

Methods, Supplementary Figures S1–S4, and Supplementary movies S1–S12.

### **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.

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### Figure captions

Figure 1: Magnetic domains at different temperatures and fields. a-e, Dynamical spin configurations of monolayer CrBr<sub>3</sub> 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<sub>3</sub>. The total strip in  $\bf{a}$  is  $400 \times 50$  nm. Only Cr atoms are showed in the honeycomb lattice arrangement of monolayer CrBr<sub>3</sub>. 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 Mz 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 CrI3. 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  $\times$  $10^9~A~cm^{-2}$ ,  $4.3\times10^9~A~cm^{-2}$ ), 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  $j = 4.3 \times 10^9 \text{ A cm}^{-2}$  are highlighted by the dashed rectangle. d, Zoom-in of the local spin-configurations at  $j = 4.3 \times 10^9 \text{ A cm}^{-2}$ ) 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 CrBr3 and CrI3, 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<sub>3</sub>. A fit using  $\nu = \mu(B - B_0)$ , where  $\mu$  is the domain wall mobility and  $B_0$  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 CrBr3 and CrI3, 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  $j_w^{CrBr_3} = 4.8 \times 10^9 \text{ A cm}^{-2}$  and  $j_w^{CrI_3} = 12.0 \times 10^9 \text{ A cm}^{-2}$ . 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  $j \le 8.5 \times 10^8$  A cm<sup>-2</sup>. e-f, Frequencies  $\omega_B$  (THz) and  $\omega_J$  (THz) of the emitted spin-waves during the domain-wall motion as a function of B and j, respectively, for CrBr<sub>3</sub> and CrI<sub>3</sub>. Dashed lines are corresponding fits to Eqs. 5-6. Similar data labelling applies for e and f.

Figure 5: Modulational instability in 2D magnets. a-c, Snapshots of the magnetisation along  $M_z$  and  $M_x$  magnetisation sation 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  $1.56 \times 10^{10}~\text{A cm}^{-2}$ .  $M_z$  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 Mz with the propagation time along a 2-\mu m ribbon of CrBr3 at a current density of  $1.56 \times 10^{10} \text{ A cm}^{-2}$ . The different line-cuts correspond to the several waveforms moving into the systems as shown in **e-g**. The dashed lines are given by  $s^+ = 5773~m~s^{-1} \pm 0.84~m~s^{-1}, \, s_1^- = 1328~m~s^{-1} \pm 1.7~m~s^{-1}$  and  $s_2^- = 882~m$  $s^{-1}\pm 1.8 \ m \ s^{-1}. \ \textbf{e-f}, Spectral \ density \ (arb. \ units) \ extracted \ from \ the \ fast \ Fourier \ transform \ of \ the \ M_z \ component \ as$ function of time in  $\mathbf{d}$  along velocities  $s^+$  and  $s_2^-$ , respectively. The characteristic frequencies of the shock waves and the domain-wall wake can be identified as  $v_{SW}$  =177.5 GHz $\pm$ 1.25 GHz and  $v_{wake}$  =6.66 $\pm$ 1.66 GHz, respectively.

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### 599 Figures

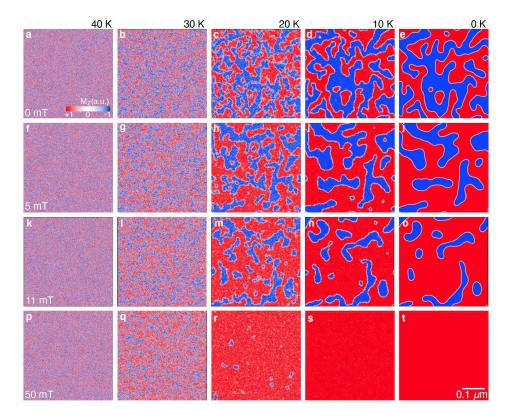


Figure 1

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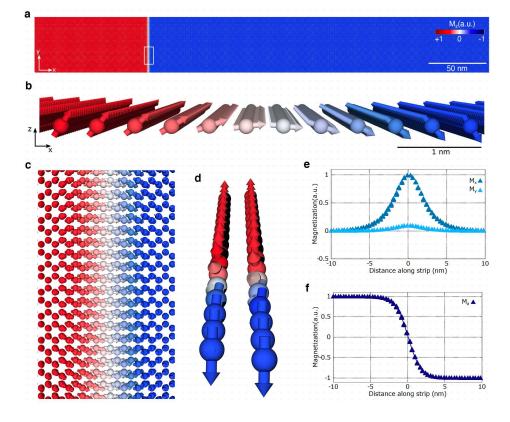


Figure 2

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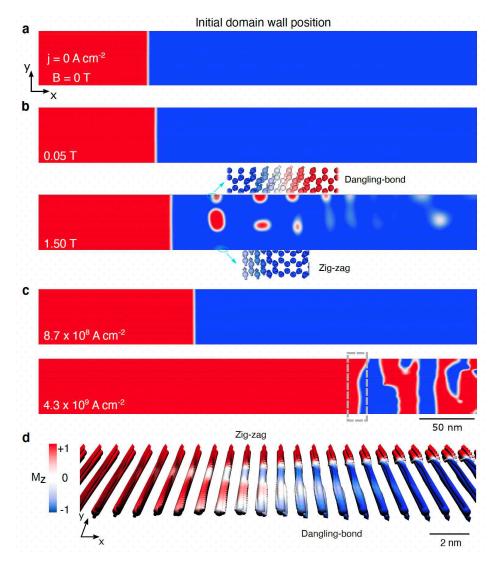


Figure 3

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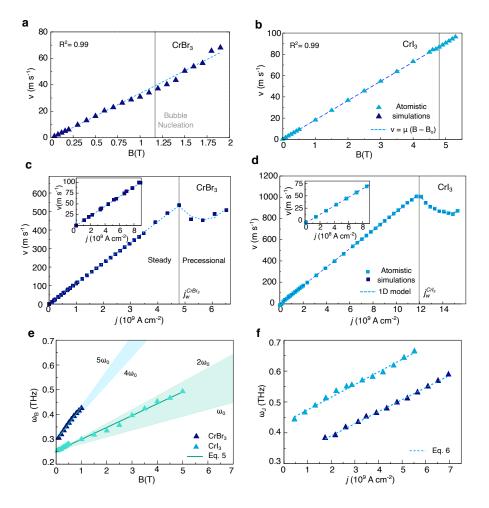


Figure 4

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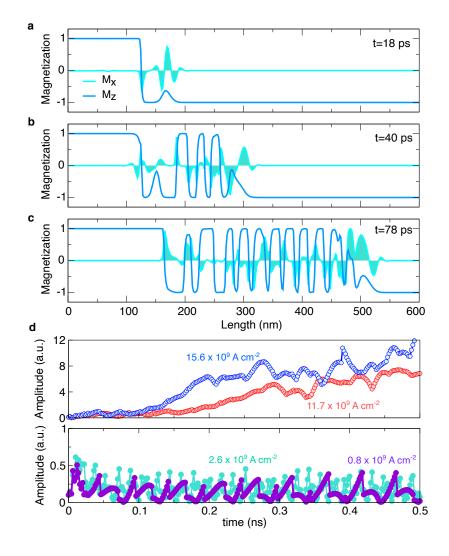


Figure 5

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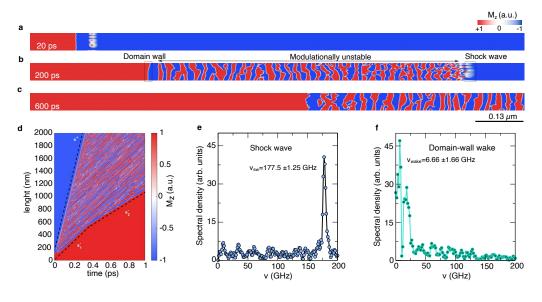


Figure 6