

# Nanopatterning reconfigurable magnetic landscapes via thermally assisted scanning probe lithography

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The search for novel tools to control magnetism at the nanoscale is crucial for the development of new paradigms in optics, electronics and spintronics. So far, the fabrication of magnetic nanostructures has been achieved mainly through irreversible structural or chemical modifications. Here, we propose a new concept for creating reconfigurable magnetic nanopatterns by crafting, at the nanoscale, the magnetic anisotropy landscape of a ferromagnetic layer exchange-coupled to an antiferromagnetic layer. By performing localized field cooling with the hot tip of a scanning probe microscope, magnetic structures, with arbitrarily oriented magnetization and tunable unidirectional anisotropy, are reversibly patterned without modifying the film chemistry and topography. This opens unforeseen possibilities for the development of novel metamaterials with finely tuned magnetic properties, such as reconfigurable magneto-plasmonic and magnonic crystals. In this context, we experimentally demonstrate spatially controlled spin wave excitation and propagation in magnetic structures patterned with the proposed method.

Metamaterials are composite materials in which new functionalities emerge from engineered combinations of carefully chosen and nanoscale designed material building blocks<sup>1–4</sup>. Among these, magnetic metamaterials allow unprecedented versatility in the design of novel device architectures, exploiting the controlled propagation of electromagnetic radiation<sup>1</sup>, surface plasmons<sup>5,6</sup> and spin waves<sup>7–9</sup>. In this context, the development of a new class of reprogrammable magnetic metamaterials, the functionality of which can be actively redesigned on demand, would open new, far-reaching directions for nanomagnetism and spintronics.

To date, the fabrication of magnetic devices through the nanopatterning of magnetic materials<sup>10–12</sup> has been carried out mainly using conventional lithography<sup>10,13</sup> or ion irradiation<sup>11,12</sup>. These techniques, in most cases, are irreversible, and their ability to finely control the modulation of the magnetic properties within the pattern is limited. Beyond these methods, advanced scanning-probe-based lithography (SPL) is gaining attention in the scientific community for its flexibility and broad applicability with emerging new materials<sup>14</sup>.

In this Article, we propose a novel, versatile approach based on SPL—‘thermally assisted magnetic scanning probe lithography’ (*tam*-SPL)—to achieve a desired and reconfigurable nanoscale engineering of the magnetic properties of a continuous film. The working principle of *tam*-SPL is the local heating and cooling of an exchange-biased ferromagnetic layer by bringing the hot tip of a scanning probe microscope (SPM) into contact with a sample, in the presence of a magnetic field. This local field cooling sets a unidirectional magnetic anisotropy in the ferromagnetic due to exchange-coupling with the adjacent antiferromagnet<sup>15</sup>. Phenomenologically, this causes a local

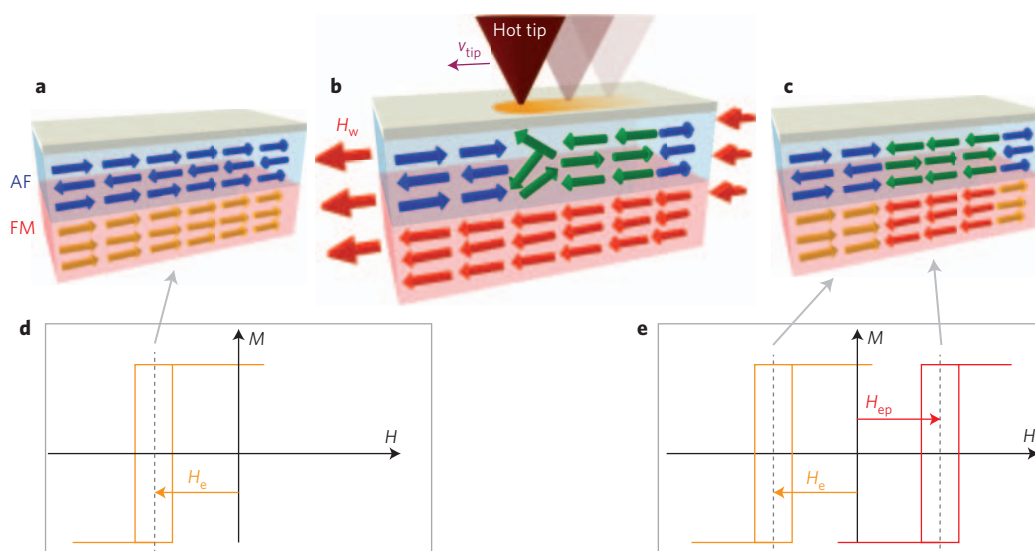
pinning of the ferromagnetic magnetization, which is observed as a shift in its hysteresis loop. Here, we demonstrate, in particular, the patterning of micro- and nanostructures with arbitrary shape, anisotropy and spin orientation. The pattern is stable at room temperature and cannot be erased permanently with magnetic fields, thus combining endurance and reconfigurability. These features can be exploited in the emerging field of magnonics to fabricate structures for the active manipulation of spin waves. As a proof-of-concept experiment, we show that local control of the spin wave excitation and propagation can be obtained in reconfigurable magnetic tracks patterned with *tam*-SPL.

## Working principle of *tam*-SPL

Magnetic patterning was performed on a Ru(2)/IrMn(7)/CoFeB(5)/SiO<sub>2</sub>(1,000)/Si (thicknesses in nm) multilayer structure, grown by magnetron sputtering, in which the IrMn/CoFeB bilayer constitutes the exchange bias system. For details of the growth and magnetic characterization of the non-patterned stack, see Methods and the Supplementary Information.

The working principle of *tam*-SPL is illustrated schematically in Fig. 1. The exchange bias system is first initialized by heating the entire sample above the blocking temperature  $T_B$  and subsequently cooling it in a uniform magnetic field  $H_i$ . This initialization sets a uniform unidirectional (UD) anisotropy axis in the ferromagnet film (Fig. 1a), with its hysteresis loop shifted by the exchange bias field  $H_e$  (Fig. 1d; see Methods for details). The hot tip of the SPM<sup>16–20</sup> is then used to locally heat the surface of the stack above  $T_B$  (160 °C) in a uniform static magnetic field  $H_w = 700$  Oe, which in this particular case is chosen to be opposite the initialization

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**Figure 1 | Magnetic patterning via *tam*-SPL.** **a**, After initialization, magnetization of the ferromagnetic (FM) layer (yellow arrows) is uniformly pinned in one direction by the exchange interaction with the antiferromagnetic (AF) layer (blue arrows). **b**, Sweeping a heated SPM tip on the sample surface in the presence of an external magnetic field  $H_w$  produces a local field cooling in the antiferromagnet (green arrows), which resets the exchange bias direction according to the underlying CoFeB spins (red arrows), aligned with  $H_w$ . **c**, When the external magnetic field  $H_w$  is removed, the magnetic domain configuration in the ferromagnet is stabilized by the local exchange bias. **d, e**, Magnetic hysteresis loops before (**d**) and after (**e**) patterning.  $H_e$  and  $H_{ep}$  indicate the opposite shift in the loops due to the exchange bias in the non-patterned and patterned areas, respectively.

1 field  $H_i$ . When the hot tip is displaced in its motion along the  
 2 writing direction, the previously heated region of the antiferromag-  
 3 net film undergoes a ‘local field cooling’ accompanied by a local  
 4 rearrangement of the spins, due to coupling with the underlying fer-  
 5 romagnet oriented in the direction of the external field,  $H_w$  (Fig. 1b).  
 6 Therefore, at the antiferromagnet–ferromagnet interface in the pat-  
 7 terned area, a different local exchange bias is established, which  
 8 shifts the local hysteresis loop to the right by a new exchange bias  
 9 field  $H_{ep}$  (Fig. 1e). On removal of the writing field  $H_w$ , the local  
 10 remanent magnetization within the ferromagnet layer is determined  
 11 by the new UD magnetic anisotropy. As a result, the spins of the fer-  
 12 romagnet in the patterned area (red arrows in Fig. 1c) are antiparal-  
 13 lel to those of the surrounding film (yellow arrows), which has  
 14 switched back to the initial state.

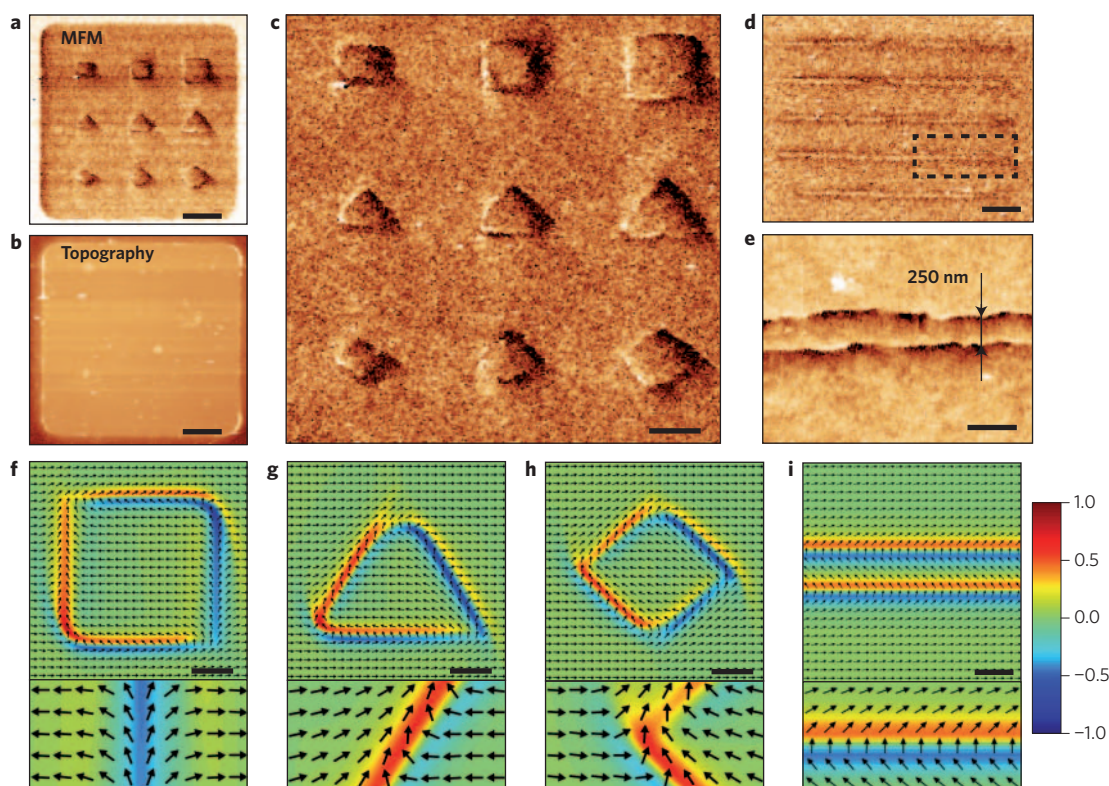
### 15 Patterning capabilities

16 The patterning capabilities of this technique are demonstrated in  
 17 Fig. 2. Square pads (sides of 20  $\mu\text{m}$ ) were first defined by optical  
 18 lithography to facilitate navigation on the sample. *tam*-SPL was  
 19 then used to define in the pad square, triangular and diamond-  
 20 shaped structures with different lateral sizes (from 1.5 to 2.5  $\mu\text{m}$ ,  
 21 Fig. 2a), with a writing speed of 3  $\mu\text{m s}^{-1}$ . The SPM heater tempera-  
 22 ture  $T_H$  was set to 560, 675 and 765  $^\circ\text{C}$  for squares, triangles and dia-  
 23 monds, respectively. These  $T_H$  values ensure that the actual  
 24 temperature at the tip–sample interface is well above  $T_B$   
 25 (Supplementary Fig. 2).  $H_w$  was set antiparallel to  $H_i$  to write struc-  
 26 tures with antiparallel remanent magnetization. For more details on  
 27 the patterning conditions, see Methods and the Supplementary  
 28 Information, where a general model for heat flow analysis developed  
 29 for thermochemical (*tc*-SPL)<sup>18</sup> has been adapted to the present case.  
 30 Figure 2a presents a magnetic force microscopy (MFM) image of a  
 31 pad, taken at zero external field (remanent state) after *tam*-SPL, and  
 32 Fig. 2b shows the topography of the same area. The opposite MFM  
 33 magnetic contrast seen at the left and right edges of the patterns  
 34 with respect to that at the pad edges demonstrates that regions  
 35 with opposite UD anisotropy and remanent magnetization have  
 36 been written by *tam*-SPL. We note that *tam*-SPL does not alter  
 37 the morphology of the films (Fig. 2b) and that the patterning is

therefore ‘purely magnetic’. A zoomed-in MFM image of the pad  
 area is shown in Fig. 2c. Although the diamonds were written at a  
 much higher temperature than the squares, the resulting magnetic  
 contrast is the same for all patterned structures, thus indicating  
 that once the blocking temperature of the system is exceeded, repro-  
 ducible patterns can be obtained without the need for precise  
 control of  $T_H$ . As the degradation of exchange bias due to Mn inter-  
 diffusion only starts to develop for temperatures above 350  $^\circ\text{C}$   
 (ref. 21), there is a wide range of film temperatures (from 160 to  
 350  $^\circ\text{C}$ ) under which patterning can be performed without  
 causing sizeable irreversibility (Fig. 4 and related discussion). The  
 fact that very demanding control of the tip temperature is not  
 required is crucial for future applications.

To gain a deeper understanding of the micromagnetic configura-  
 tion giving rise to the magnetic contrast observed with MFM, we  
 performed micromagnetic simulations (see Supplementary  
 Information) of the square, triangle and diamond patterns, as  
 shown in Fig. 2f–h, respectively, where the black arrows indicate  
 the orientation of the spins within the CoFeB film. The simulated  
 out-of-plane component of the force on the ferromagnetic tip is pre-  
 sented in Fig. 2f–h using a blue–red (attractive–repulsive) colour  
 code that can be associated with the dark–white contrast of MFM  
 (see Supplementary Information for details). The agreement  
 between simulations and experiments is very good for all patterns.  
 It is worth noting that different kinds of 180 $^\circ$  domain wall can be  
 engineered by controlling the pattern shape. The fine details of  
 the domain wall structure for each pattern are illustrated in the  
 simulations in Fig. 2f–i, which show that by choosing the angle of  
 the pattern side with respect to  $H_i$  and  $H_w$ , it is possible to tune  
 the domain wall magnetic configuration.

The determination of the minimum feature size achievable by  
 performing *tam*-SPL in our multilayer system was assessed by  
 writing single lines with  $H_w$  antiparallel to  $H_i$  and with a spacing  
 of 2  $\mu\text{m}$  (Fig. 2d). The high-resolution MFM image in Fig. 2e of  
 the dashed area in Fig. 2d shows that the average width of a single  
 line, measured over a 2  $\mu\text{m}$  section, is 300 nm, with a minimum  
 of 250 nm. This demonstrates that, in this condition, the mini-  
 mum spacing between lines during writing (allowing them to



**Figure 2 | MFM characterization and micromagnetic simulations of the patterned domain structures.** **a,b**, MFM image (**a**) and topography (**b**) of the same pad after performing *tam*-SPL. The patterned domains are clearly visible in the MFM image, but no change is observed in the surface topography. **c**, Zoomed-in MFM image of square (top row), triangular (centre row) and diamond-shaped (bottom row) magnetic domains with dimensions of 2.5  $\mu\text{m}$  (right column), 2  $\mu\text{m}$  (centre column) and 1.5  $\mu\text{m}$  (left column). The magnetic contrast marks the domain boundaries. **d**, MFM of single lines (12  $\mu\text{m}$  long). **e**, High-resolution MFM image of the dashed rectangle in **d**, showing a minimum width of 250 nm. All MFM images were acquired under no external magnetic field. **f-i**, Micromagnetic simulation of the magnetic configuration of the patterned domains. Arrows indicate the direction of the spins, and the colour map shows the normalized magnetic force acting on the MFM tip. Bottom panels: zoomed-in view of the domain walls. In square structures (**f**), head-to-head and tail-to-tail domain walls are defined on the left and right sides, respectively, with the spins within the domain wall aligned along the wall. A different micromagnetic configuration is observed on the top and bottom sides, where the domain wall spins are perpendicular to the wall. In the triangle and diamond shapes (**g,h**), the arrangement of the spins within the wall is between the two cases described above. Scale bars: 4  $\mu\text{m}$  (**a,b**), 2  $\mu\text{m}$  (**c,d**), 500 nm (**e**), 1  $\mu\text{m}$  (**f-h**), 250 nm (**i**).

Q5 1 keep their individuality) is on the order of 300 nm. The patterned  
 2 lines present two domain walls of the same topology as those  
 3 observed in the horizontal sides of the squares (Fig. 2f,i), with mag-  
 4 netization antiparallel with respect to the non-patterned area. The  
 5 minimum feature size and the corrugation of the lines are deter-  
 6 mined both by the domain wall width, which depends on the micro-  
 7 magnetic properties of the system, and by the spatial profile of the  
 8 exchange bias written by *tam*-SPL. Micromagnetic simulations indi-  
 9 cate that the width of ideal 180° domain walls, with magnetization  
 10 parallel to the wall far away from it, is on the order of 350 nm, in  
 11 good agreement with the experimental width of the lines in  
 12 Fig. 2e (Supplementary Fig. 8 and ref. 22). The spatial profile of  
 13 the exchange bias is given by the finite width of the thermal  
 14 profile produced by the hot tip in the antiferromagnet layer, as  
 15 well as by the spatial distribution of the blocking temperatures of  
 16 the system, which reflects the distribution of grain sizes in the anti-  
 17 ferromagnet layer (see Supplementary Information). An improved  
 18 spatial resolution and a reduced line corrugation can thus be  
 19 achieved by optimizing the patterning conditions, the micromag-  
 20 netic properties of the system<sup>22</sup> (for example, the exchange bias  
 21 strength, magnetocrystalline anisotropy and exchange stiffness), the  
 22 grain size distribution and the thermal properties of the materials<sup>23</sup>.  
 23 Regarding the spatial confinement of the thermal profile, it has  
 24 already been demonstrated that by performing *tc*-SPL on thermally  
 25 insulating films<sup>18</sup>, a 15 nm feature size can be achieved, which is at

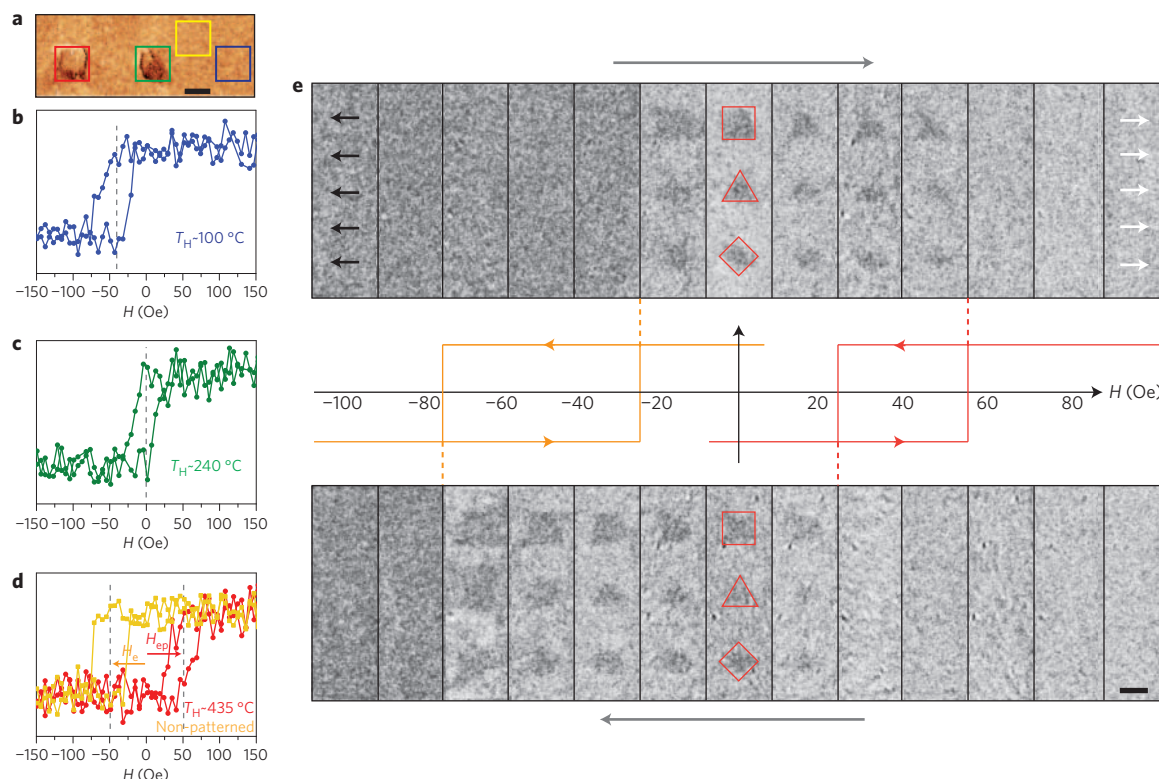
least comparable with state-of-the-art near-field transducers for  
 magnetic recording technology<sup>24</sup>. For the writing speed, note that  
 the crucial parameter is the time required to locally heat the sample  
 above  $T_B$ . Further improvement, that is, down to 1  $\mu\text{s}$  per point  
 feature, could be obtained with a different thermal cantilever design<sup>25</sup>.

In perspective, well-developed local heating technologies like  
 those used in magneto-optical recording (MOR) and heat-assisted  
 magnetic recording (HAMR) could be exploited for scaling up the  
 presented approach<sup>26–28</sup>.

### Tunability and reconfigurability

Figure 3 shows that it is possible to finely tune the patterned mag-  
 netic anisotropy landscape by controlling the tip temperature.  
 Figure 3a presents an MFM image taken in the remanent state for  
 square areas patterned at  $T_H = 100$ , 240 and 435 °C (right to left  
 in the figure) while applying  $H_w = 700$  Oe antiparallel to  $H_i$ .  
 When  $T_H = 100$  °C, the local temperature of the sample is well  
 below the blocking temperature, so no magnetic contrast is seen  
 (blue square). Correspondingly, the shift in the local hysteresis  
 loop measured with magneto-optical Kerr effect microscopy  
 ( $\mu$ -MOKE) along the UD anisotropy direction (Fig. 3b), approxi-  
 mately –50 Oe, is comparable to the shift of the loop (yellow loop  
 in Fig. 3d) measured in the adjacent non-patterned area (yellow  
 square in Fig. 3a). The patterned area written with  $T_H = 240$  °C  
 shows magnetic contrast, and the corresponding

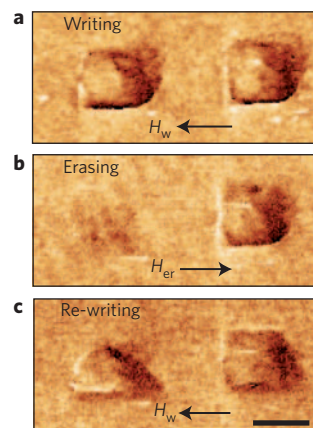




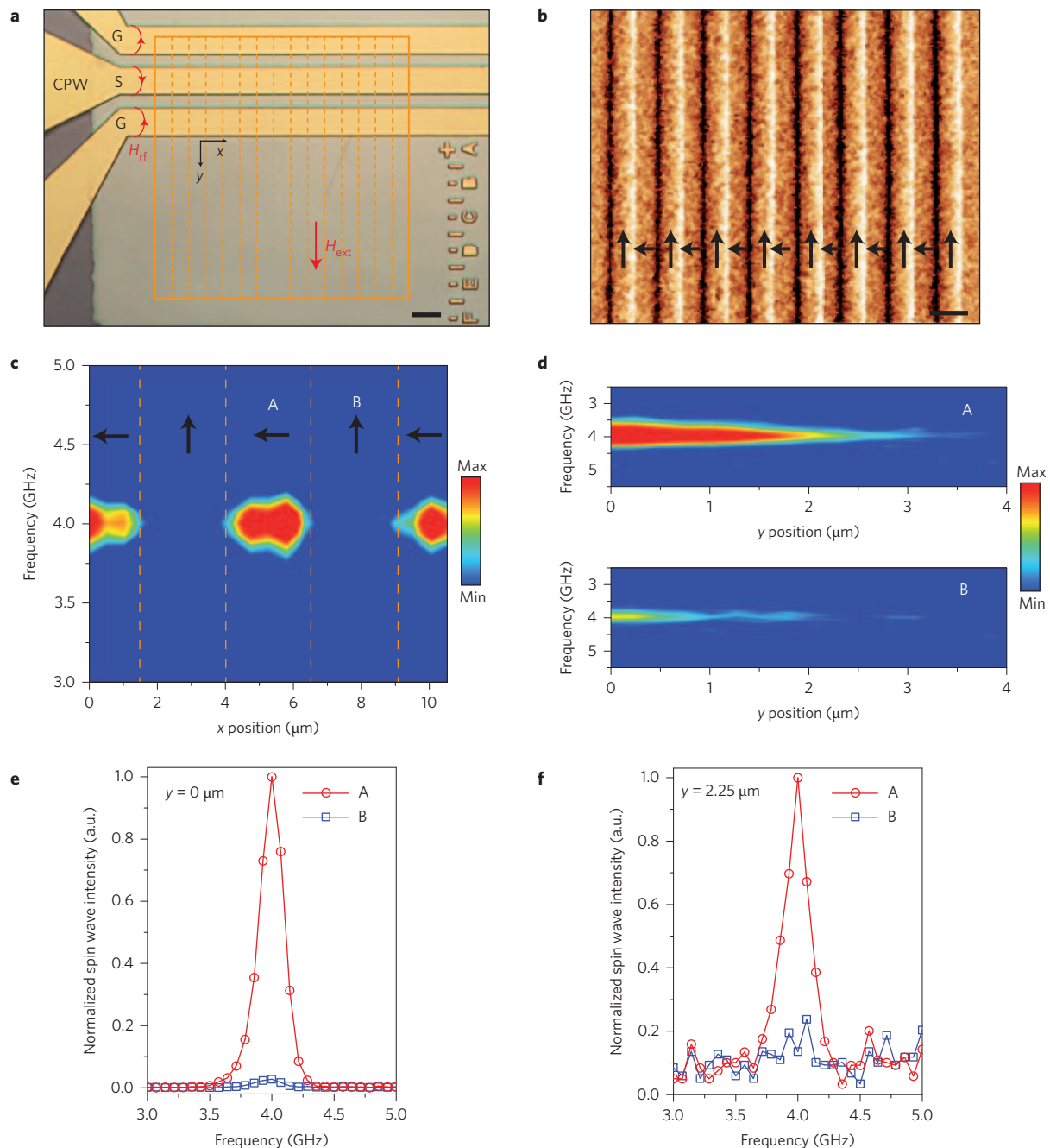
**Figure 3 | Tunability of magnetic anisotropies and evolution of patterned domains with external magnetic field.** **a**, MFM image at zero magnetic field from a non-patterned area (yellow square) and from areas patterned with  $T_H = 100$  °C (blue square), 240 °C (green square) and 435 °C (red square), and  $H_w = 700$  Oe. **b–d**,  $\mu$ -MOKE hysteresis loops corresponding to the blue, green, and red and yellow squares in **a**, respectively. Starting from the initialized sample (yellow loop,  $H_e = -50$  Oe), by increasing the heater temperature it is possible to tune the magnetic anisotropy and hence the exchange bias field (from  $H_{ep} = -50$  Oe, blue loop; up to  $H_{ep} = +50$  Oe, red loop). **e**,  $\mu$ -MOKE frames from a movie acquired while sweeping a magnetic field parallel to the UD anisotropy axis, from  $-100$  to  $+100$  Oe (top sequence) and from  $+100$  to  $-100$  Oe (bottom sequence). At the beginning of the top sequence, the magnetization of both patterned and non-patterned areas lies in the negative direction (dark contrast); at around  $-20$  Oe, the magnetization of the non-patterned film switches, giving rise to a bright contrast, while the patterned areas remain dark. The dark contrast eventually disappears at about  $+50$  Oe, when the switching takes place also in the patterned areas. An analogue switching behaviour is observed in the bottom sequence. In the centre, the hysteresis loops from the non-patterned (yellow) and patterned (red) areas are sketched and the switching fields are marked. Scale bars,  $2\ \mu\text{m}$  (**a,e**).

1 local hysteresis loop (Fig. 3c) is centred at zero magnetic field. By  
 2 increasing  $T_H$  to 435 °C (red square in Fig. 3a), the local hysteresis  
 3 loop is eventually shifted to the right by  $+50$  Oe (Fig. 3d), showing  
 4 full inversion of the exchange bias field. The zero shift of the loop at  
 5 240 °C results from an incomplete exchange bias inversion due to  
 6 the expected distribution of  $T_B$  in the film arising from the presence  
 7 of grains with different size<sup>15,29</sup> (Supplementary Fig. 1). This  
 8 demonstrates the possibility to pattern complex magnetic aniso-  
 9 tropy landscapes in a continuous multilayer by tuning the heater  
 10 temperature during writing. Moreover, by setting  $H_w$  at different  
 11 angles with respect to  $H_i$ , areas can be patterned with the UD anisotropy axis arbitrarily oriented with respect to the film UD anisotropy set during initialization (Supplementary Fig. 3).

14 Analysis of the magnetic behaviour of the patterned area under  
 15 an applied magnetic field, including the stability of the patterns,  
 16 was carried out by  $\mu$ -MOKE. Figure 3e presents some frames  
 17 from a movie (Supplementary Movie 1), which show the field-  
 18 induced evolution of the magnetization in the patterned area of  
 19 Fig. 2c (right column). The appearance (disappearance) of the  
 20 pattern is in agreement with the expected hysteresis loops of the pat-  
 21 terned (non-patterned) regions, sketched in red (yellow).  
 22 Furthermore—and crucial for technological applications—we have  
 23 verified that the same remanent state (red shapes in Fig. 3e) is  
 24 always recovered, even after 2,000 magnetization switching cycles  
 25 performed with fields up to 1,000 Oe (data not shown), and that  
 26 the patterns are stable six months after writing. Indeed, the



**Figure 4 | Writing-erasing-rewriting capability.** **a**, MFM image of two squares patterned with  $T_H = 600$  °C and  $H_w = 700$  Oe. **b**, The same area after erasure of the left square obtained by scanning it with  $T_H = 600$  °C and an erasing field of  $H_{er} = -700$  Oe, opposite to  $H_w$  and in the same direction as the initializing field  $H_i$ . **c**, A triangle is then rewritten on the left area with  $T_H = 470$  °C and  $H_w = 700$  Oe. Note that both the erasure and rewriting processes leave the right square unaffected. Scale bar,  $2\ \mu\text{m}$ .



**Figure 5 | Patterning magnonic structures.** **a**, waveguide (CPW) generating the Oersted field  $H_{rf}$  and the pad where the *tam*-SPL patterning was performed. The orange rectangle marks the patterned area. Scale bar, 4  $\mu\text{m}$ . **b**, MFM image of the magnonic structures, consisting of 2.5- $\mu\text{m}$ -wide tracks with alternating  $0^\circ$  and  $90^\circ$  oriented remanent static magnetization (black arrows). The track boundaries are marked by dashed lines in **a**. Scale bar, 4  $\mu\text{m}$ . **c**, One-dimensional map of the spin wave intensity measured at remanence by  $\mu$ -BLS along the  $x$  direction (with a 0.5  $\mu\text{m}$  step size), immediately outside the CPW. Spin waves are efficiently excited within tracks of type A. **d**, One-dimensional map of the spin wave intensity measured at remanence by  $\mu$ -BLS as a function of distance  $y$  from the CPW (with a 0.25  $\mu\text{m}$  step size). **e, f**, Spin wave spectra measured in track A (red circles) and track B (blue squares) at distances of  $y = 0 \mu\text{m}$  (**e**) and  $y = 2.25 \mu\text{m}$  (**f**) from the CPW.

1 resilience of the *tam*-SPL patterns is derived from the stability of the  
2 exchange bias, a key property that is also exploited in magnetic  
3 memory applications<sup>30–33</sup>. This feature can be used to implement  
4 a permanent functionality, similar to the case where magnetic  
5 devices are patterned by conventional techniques such as lithogra-  
6 phy and ion irradiation.

7 The reversibility of *tam*-SPL is illustrated in Fig. 4. Figure 4a  
8 shows a MFM image, under zero external field, of two squares pat-  
9 terned with  $T_H = 600^\circ\text{C}$  and  $H_w = 700 \text{ Oe}$  antiparallel to  $H_i$ .

Figure 4b shows the same area after local cancellation of the left  
square, achieved by scanning the same area with  $T_H = 600^\circ\text{C}$  in  
an erasing field of  $H_{er} = -700 \text{ Oe}$ , opposite to  $H_w$ . The magnetic  
contrast almost completely disappears in the left square, but  
remains unperturbed in the right, indicating that the pattern has  
been selectively erased. Figure 4c shows that it is possible to  
rewrite the erased area with another shape, a triangle in this case,  
with  $T_H = 600^\circ\text{C}$  and  $H_w = 700 \text{ Oe}$ . The pattern can also be com-  
pletely erased by performing uniform field cooling, thus providing full

1 flexibility in writing and reversibly configuring the magnetic  
2 anisotropy landscape.

### 3 Magnonic applications

4 Here, we present a proof-of-concept experiment, performed by  
5 micro-focused Brillouin light scattering ( $\mu$ -BLS), showing the  
6 potential applications of *tam*-SPL in the field of magnonics<sup>34–38</sup>.  
7 So far, magnonic devices have been fabricated by the physical pat-  
8 terning of magnetic thin films<sup>39</sup>, modulating their thickness<sup>40</sup> or  
9 (as in the case of bi-component magnetic devices) creating a peri-  
10 odic array made of two magnetic materials<sup>41</sup>. Here, we propose to  
11 use *tam*-SPL to pattern magnonic structures in a continuous film,  
12 thus avoiding the scattering at physical interfaces created by stan-  
13 dard lithography and providing an easy way to introduce reconfigur-  
14 ability and tunability into magnonics.

15 The investigated sample consisted of a  $60\text{ }\mu\text{m} \times 60\text{ }\mu\text{m}$  square  
16 pad, defined by optical lithography on the same multilayer structure  
17 in Fig. 2. *tam*-SPL was performed by applying  $H_w$  perpendicular to  
18  $H_i$  to define  $2.5\text{-}\mu\text{m}$ -wide tracks with alternating  $0^\circ$  and  $90^\circ$  oriented  
19 remanent magnetization over a total area of  $38\text{ }\mu\text{m} \times 39\text{ }\mu\text{m}$  (orange  
20 rectangle in Fig. 5a). The MFM image of the pattern at remanence is  
21 presented in Fig. 5b, where the black arrows indicate the magnetiza-  
22 tion direction within the pattern. The sharp transition between the  
23 two different magnetization configurations is marked by well-  
24 defined and sharp  $90^\circ$  domain walls.

25 Coplanar wave guides (CPWs), with a ground-to-ground dis-  
26 tance of  $12\text{ }\mu\text{m}$  and oriented perpendicular to the longitudinal  
27 direction of the *tam*-SPL tracks, were fabricated by electron-beam  
28 lithography and liftoff.  $\mu$ -BLS measurements were then performed  
29 to map the spin wave intensity distribution.

30 Figure 5c shows the colour-coded spin wave intensity on a logar-  
31 ithmic scale measured at remanence, in the 3–5 GHz frequency  
32 range, as a function of the  $x$  position along the CPW arms. The  
33 BLS linear scan was performed just outside the CPW, at  $y=0$  in  
34 the reference system of Fig. 5a. The orange dashed lines mark the  
35 boundaries of the tracks, while the black arrows indicate the local  
36 direction of the static magnetization, which is parallel (perpendic-  
37 lar) with respect to the CPW in type A (B) tracks. Remarkably, pro-  
38 pagating spin waves are efficiently excited only in type A tracks,  
39 whereas the spin waves' excitation efficiency is strongly reduced in  
40 type B tracks. A direct comparison of BLS spectra acquired just  
41 outside the CPW (Fig. 5e) shows that the spin wave intensity is  
42 about 36 times higher in the type A track (red circles) than in  
43 type B (blue squares).

44 Such behaviour can be explained by considering that, for this  
45 CPW geometry, spin waves are efficiently excited mainly by the  
46 coupling between the in-plane component of the microwave field  
47 ( $H_{rf}$  in Fig. 5a) and the corresponding component of the dynamical  
48 magnetization, associated with the spin waves, while the out-of-  
49 plane component of the microwave field plays a minor role.  
50 Within the pattern, the efficiency of the coupling is high in type  
51 A and low in type B tracks, depending on whether the in-plane com-  
52 ponent of the microwave field and the corresponding component of  
53 the dynamic magnetization are parallel or perpendicular<sup>42,43</sup>.

54 Figure 5d shows the colour-coded spin wave intensity on a log-  
55 arithmic scale acquired at remanence as a function of distance  $y$   
56 from the CPW, in the centre of two adjacent tracks. We found  
57 that spin waves attenuate rapidly in the type B track (Fig. 5d,  
58 bottom), but in the type A track (Fig. 5d, top) they propagate  
59 efficiently over a distance comparable to that observed in Py  
60 and CoFe waveguides prepared by conventional lithographic  
61 techniques<sup>44,45</sup>. This result is confirmed by the BLS spectra acquired  
62 at  $y=2.25\text{ }\mu\text{m}$  in tracks A and B (Fig. 5f). This finding can be  
63 explained taking into account that for perpendicular alignment of  
64 the spin waves wavevector and local magnetization, the group vel-  
65 ocity of the spin waves is much larger than for parallel alignment<sup>46</sup>.

The above result indicates that it is possible to locally control the  
spin wave excitation and propagation by creating remanent magne-  
tization patterns via *tam*-SPL. This marks an initial step for the  
implementation of *tam*-SPL-based tunable and reconfigurable  
magnonic devices.

Supplementary Fig. 7 demonstrates the design of phase shifters  
and magnonic crystals exploiting the modulation of the refractive  
index seen by the spin waves in patterned regions with different rela-  
tive orientation of the static magnetization and of the spin waves'  
wavevector. A single stripe can be used to implement a phase  
shifter to be inserted in Mach–Zehnder-type interferometers,  
which have been proposed as basic building blocks for computing<sup>47</sup>.  
In addition, a periodic arrangement of stripes such as those shown  
in Fig. 2d can be used to realize a magnonic crystal showing the  
emergence of characteristic bandgaps. The bandwidth and position  
of the bandgaps can be tuned by controlling the stripe width, their  
separation and the width of the regions of spin rotation between the  
stripes. Because all these parameters can be set reversibly during  
patterning, our technique allows the fabrication of fully reconfigur-  
able magnonic crystals and devices. Moreover, active tuning of the  
magnonic properties after patterning can be achieved by controlling  
the domain extension and magnetization orientation via external  
magnetic fields (Fig. 3).

### Conclusions

In this Article, we have introduced *tam*-SPL, a straightforward,  
single-step method for nanopatterning reprogrammable magnetic  
metamaterials, leaving the film chemistry and topography unper-  
turbed. In particular, we have demonstrated the patterning of arbi-  
trarily shaped magnetic micro- and nanostructures, with size down  
to  $250\text{ nm}$  and remanent magnetization along an arbitrarily chosen  
direction, which are reconfigurable and stable against external mag-  
netic perturbations. In perspective, further improvement of the  
spatial resolution could be achieved by optimizing the patterning  
parameters as well as the micromagnetic properties, thermal con-  
ductivity and granularity of the magnetic multilayer. A higher pat-  
terning speed could be obtained by using arrays of many heatable  
cantilevers<sup>48</sup> or using near-field transducers, allowing for highly  
localized and fast heating, with theoretical spot sizes down to  
 $35\text{ nm}$  (ref. 24).

In addition, due to the extreme versatility of SPL, the magnetic  
patterning can be extended from continuous films to physically  
micro- and nanopatterned supports. Novel architectures for mag-  
netic computing and spintronics can be envisaged, where repro-  
grammable magnetic metamaterials control the propagation of  
quasiparticles, such as domain walls in magnetic circuits, or spin  
waves and surface plasmons in magneto-plasmonic crystals. In par-  
ticular, we have demonstrated local control of spin wave excitation  
and propagation in magnetic tracks patterned by *tam*-SPL. This  
result paves the way to the realization of reconfigurable and actively  
tunable devices, such as spin wave phase shifters and filters based on  
magnonic crystals.

### Methods

Methods and any associated references are available in the online  
version of the paper.

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### References

- Schurig, D. *et al.* Metamaterial electromagnetic cloak at microwave frequencies. *Science* **314**, 977–980 (2006).
- Zheludev, N. I. & Kivshar, Y. S. From metamaterials to metadevices. *Nature Mater.* **11**, 917–924 (2012).
- Nikitov, S. a., Tailhades, P. & Tsai, C. S. Spin waves in periodic magnetic structures—magnonic crystals. *J. Magn. Magn. Mater.* **236**, 320–330 (2001).



4. Silva, A. *et al.* Performing mathematical operations with metamaterials. *Science* **343**, 160–164 (2014).
5. Temnov, V. V. Ultrafast acousto-magneto-plasmonics. *Nature Photon.* **6**, 728–736 (2012).
6. Maccaferri, N. *et al.* Resonant enhancement of magneto-optical activity induced by surface plasmon polariton modes coupling in 2D magnetoplasmonic crystals. *ACS Photon.* **2**, 1769–1779 (2015).
7. Kobljanskij, Y. *et al.* Nano-structured magnetic metamaterial with enhanced nonlinear properties. *Sci. Rep.* **2**, 478 (2012).
8. Lenk, B., Ulrichs, H., Garbs, F. & Münzenberg, M. The building blocks of magnonics. *Phys. Rep.* **507**, 107–136 (2011).
9. Chumak, A. V., Vasyuchka, V. I., Serga, A. A. & Hillebrands, B. Magnon spintronics. *Nature Phys.* **11**, 453–461 (2015).
10. Monticelli, M. *et al.* On-chip magnetic platform for single-particle manipulation with integrated electrical feedback. *Small* <http://dx.doi.org/10.1002/smll.201500916> (2015).
11. Chappert, C. Planar patterned magnetic media obtained by ion irradiation. *Science* **280**, 1919–1922 (1998).
12. Kim, S. *et al.* Nanoscale patterning of complex magnetic nanostructures by reduction with low-energy protons. *Nature Nanotech.* **7**, 567–571 (2012).
13. Ross, C. A. Patterned magnetic recording media. *Annu. Rev. Mater. Res.* **31**, 203–235 (2001).
14. Garcia, R., Knoll, A. W. & Riedo, E. Advanced scanning probe lithography. *Nature Nanotech.* **9**, 577–587 (2014).
15. Nogués, J. & Schuller, I. K. Exchange bias. *J. Magn. Magn. Mater.* **192**, 203–232 (1999).
16. Szoszkiewicz, R. *et al.* High-speed, sub-15 nm feature size thermochemical nanolithography. *Nano Lett.* **7**, 1064–1069 (2007).
17. Wang, D. *et al.* Thermochemical nanolithography of multifunctional nanotemplates for assembling nano-objects. *Adv. Funct. Mater.* **19**, 3696–3702 (2009).
18. Wei, Z. *et al.* Nanoscale tunable reduction of graphene oxide for graphene electronics. *Science* **328**, 1373–1376 (2010).
19. Kim, S. *et al.* Direct fabrication of arbitrary-shaped ferroelectric nanostructures on plastic, glass, and silicon substrates. *Adv. Mater.* **23**, 3786–3790 (2011).
20. Pires, D. *et al.* Nanoscale three-dimensional patterning of molecular resists by scanning probes. *Science* **328**, 732–735 (2010).
21. Fernandez-Outon, L. E., Araújo Filho, M. S., Araújo, R. E., Ardisson, J. D. & Macedo, W. A. A. Setting temperature effect in polycrystalline exchange-biased IrMn/CoFe bilayers. *J. Appl. Phys.* **113**, 17D704 (2013).
22. Albisetti, E. & Petti, D. Domain wall engineering through exchange bias. *J. Magn. Magn. Mater.* **400**, 230–235 (2016).
23. Algré, E., Gaudin, G., Bsiesy, A. & Nozières, J. Improved patterned media for probe-based HAMR. *IEEE Trans. Magn.* **41**, 2857–2859 (2005).
24. Miao, L., Stoddart, P. R. & Hsiang, T. Y. Novel aluminum near field transducer and highly integrated micro-nano-optics design for heat-assisted ultra-high-density magnetic recording. *Nanotechnology* **25**, 295202 (2014).
25. King, W. P., Bhatia, B., Felts, J. R., Kim, H. J. & Kwon, B. Heated atomic force microscope cantilevers and their applications. *Annu. Rev. Heat Transf.* **XVI**, 287–326 (2013).
26. Wu, A. Q. *et al.* HAMR areal density demonstration of 1 + Tbpsi on spindisk. *IEEE Trans. Magn.* **49**, 779–782 (2013).
27. Saito, J., Sato, M., Matsumoto, H. & Akasaka, H. Direct overwrite by light power modulation on magneto-optical multi-layered media. *Jpn J. Appl. Phys.* **26**, 155–159 (1987).
28. Stanciu, C. *et al.* All-optical magnetic recording with circularly polarized light. *Phys. Rev. Lett.* **99**, 047601 (2007).
29. O'Grady, K., Fernandez-Outon, L. E. & Vallejo-Fernandez, G. A new paradigm for exchange bias in polycrystalline thin films. *J. Magn. Magn. Mater.* **322**, 883–899 (2010).
30. Petti, D. *et al.* Storing magnetic information in IrMn/MgO/Ta tunnel junctions via field-cooling. *Appl. Phys. Lett.* **102**, 192404 (2013).
31. Marti, X. *et al.* Room-temperature antiferromagnetic memory resistor. *Nature Mater.* **13**, 367–374 (2014).
32. Prejbeanu, I. L. *et al.* Thermally assisted MRAMs: ultimate scalability and logic functionalities. *J. Phys. D* **46**, 074002 (2013).
33. Papusoi, C. *et al.* Reversing exchange bias in thermally assisted magnetic random access memory cell by electric current heating pulses. *J. Appl. Phys.* **104**, 013915 (2008).
34. Camley, R. E. *et al.* High-frequency signal processing using magnetic layered structures. *J. Magn. Magn. Mater.* **321**, 2048–2054 (2009).
35. Kim, S.-K., Lee, K.-S. & Han, D.-S. A gigahertz-range spin-wave filter composed of width-modulated nanostrip magnonic-crystal waveguides. *Appl. Phys. Lett.* **95**, 082507 (2009).
36. Chumak, A. V., Serga, A. A. & Hillebrands, B. Magnon transistor for all-magnon data processing. *Nature Commun.* **5**, 4700 (2014).
37. Krawczyk, M. & Grundler, D. Review and prospects of magnonic crystals and devices with reprogrammable band structure. *J. Phys. Condens. Matter* **26**, 123202 (2014).
38. Csaba, G., Papp, A. & Porod, W. Spin-wave based realization of optical computing primitives. *J. Appl. Phys.* **115**, 17C741 (2014).
39. Neusser, S. & Grundler, D. Magnonics: spin waves on the nanoscale. *Adv. Mater.* **21**, 2927–2932 (2009).
40. Gubbiotti, G. *et al.* Collective spin waves on a nanowire array with step-modulated thickness. *J. Phys. D* **47**, 105003 (2014).
41. Tacchi, S. *et al.* Forbidden band gaps in the spin-wave spectrum of a two-dimensional bicomponent magnonic crystal. *Phys. Rev. Lett.* **109**, 137202 (2012).
42. Kalinikos, B. A. Excitation of propagating spin waves in ferromagnetic films. *IEEE Proc. H* **127**, 4 (1980).
43. Brächer, T. *et al.* Generation of propagating backward volume spin waves by phase-sensitive mode conversion in two-dimensional microstructures. *Appl. Phys. Lett.* **102**, 2011–2016 (2013).
44. Vogt, K. *et al.* Realization of a spin-wave multiplexer. *Nature Commun.* **5**, 3727 (2014).
45. Urazhdin, S. *et al.* Nanomagnonic devices based on the spin-transfer torque. *Nature Nanotech.* **9**, 509–513 (2014).
46. Stancil, D. D. & Prabhakar, A. *Spin Waves Theory and Applications* (Springer, 2009).
47. Schneider, T. *et al.* Realization of spin-wave logic gates. *Appl. Phys. Lett.* **92**, 022505 (2008).
48. Carroll, K. M. *et al.* Parallelization of thermochemical nanolithography. *Nanoscale* **6**, 1299–1304 (2014).

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## Author contributions

E.A., with the help of D.P., conceived and designed the experiments. E.R. and R.B. coordinated and supervised the research. E.A. performed patterning experiments, MFM characterization and simulations. D.P. fabricated the samples. W.P.K. provided the thermal SPM tips. E.A., M.P., P.V. and R.B. performed MOKE characterization. E.A. and D.P. fabricated the samples for  $\mu$ -BLS measurements. M.M. and S.T. performed  $\mu$ -BLS measurements. A.P., G.C. and W.P. performed the simulation of the magnonic structures. E.A., D.P., M.M., S.T., P.V., E.R. and R.B. wrote the manuscript. All authors contributed to discussions regarding the research.

## Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at [www.nature.com/reprints](http://www.nature.com/reprints). Correspondence and requests for materials should be addressed to E.A., E.R. and R.B.

## Competing financial interests

The authors declare no competing financial interests.

## Methods

**Sample fabrication and characterization.**  $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$  (5 nm)/ $\text{Ir}_{22}\text{Mn}_{78}$  (7 nm)/Ru (2 nm) stacks were deposited on  $\text{Si}/\text{SiO}_2$  (1,000 nm) substrates by d.c. magnetron sputtering using an AJA Orion8 system with a base pressure below  $1 \times 10^{-8}$  torr. During deposition, a 300 Oe magnetic field ( $H_G$ ) was applied in the sample plane to set the magnetocrystalline uniaxial (UA) anisotropy direction in the CoFeB layer and the exchange bias direction in the as-grown sample. Both the critical temperature at which the exchange bias disappears (blocking temperature  $T_B$ ) and the exchange bias strength decrease with antiferromagnetic layer (IrMn) thickness. We used a 7 nm IrMn layer, as this thickness ensures both a strong exchange bias and a  $T_B$  above room temperature (160 °C, as shown in Supplementary Fig. 1), allowing local field cooling to be performed by scanning the heatable SPM tip. For the ferromagnet, the 5 nm CoFeB thickness ensures both a sizeable exchange bias, which decreases for higher ferromagnet thickness<sup>15</sup>, and a straightforward characterization of the magnetic domains by MFM and  $\mu$ -MOKE. The multilayer surface roughness measured in a  $1 \mu\text{m}^2$  area is 0.15 nm (root mean square). Squared pads with a  $400 \mu\text{m}^2$  area were microfabricated by optical lithography and ion milling, allowing for the individuation of the magnetic patterns defined by *tam*-SPL. After microfabrication, to set the UD anisotropy axis in the CoFeB film, the samples underwent field cooling starting at 220 °C (above blocking temperature  $T_B$ ) in vacuum and in a 4,000 Oe magnetic field  $H_i$  applied along the UA axis of the CoFeB film. The magnetic characterization of the stacks was performed in a Microsense vibrating sample magnetometer equipped with a heater for temperature-dependent measurements.

***tam*-SPL.** *tam*-SPL was performed with a modified Agilent 5500 SPM system equipped with custom-made silicon SPM cantilevers integrated with a Joule-heating resistive heater<sup>17,18,25</sup>. Details about these thermal cantilevers and temperature calibration are provided in the Supplementary Information. A National Instruments NI cDAQ-9178 was used to control and record the current passing through the cantilever to achieve Joule heating. To pattern arbitrary one- or two-dimensional geometries, such as lines, polygons and complex planar shapes, MATLAB scripts were used in combination with Agilent PicoView software. The magnetic patterns were written by raster-scanning the heated tip in contact mode using a linear speed of  $3 \mu\text{m s}^{-1}$ . Two permanent magnets were used to generate the uniform 700 Oe external magnetic field applied in the sample plane during patterning. An arbitrary angle between the direction of the external field and the initialization direction of the sample was set by physically rotating the sample with respect to the magnets.

**MFM characterization.** MFM characterization of the patterned magnetic domains was performed with the same Agilent 5500 SPM system used for *tam*-SPL, but equipped with a Nanosensors PPP-MFMR AFM magnetic probe. MFM imaging was performed in lift mode, with lift heights of 100 nm (Figs 2a,c,d, 3a, 4 and 5b and Supplementary Figs 3 and 4) and 30 nm (Fig. 2e).

**$\mu$ -MOKE characterization.** To conduct investigations of the magnetization distribution at the sub-micrometre length scale we used a characterization tool based on magneto-optic Kerr effect imaging/magnetometry based on an optical wide-field polarization microscope optimized for Kerr microscopy (Evico). Arbitrary magnetic fields can be applied during observation so that domain nucleation and magnetization processes can be observed. Indeed, the domain contrast in the image is directly sensitive to the projection of the magnetization along the sensitivity direction, which is set either parallel or perpendicular to the externally applied field. For this purpose, the microscope was equipped with a rotatable bipolar electromagnet that allowed the application of a magnetic field with intensity up to 5,000 Oe along an arbitrary direction in the sample plane. Two different operational modes of the microscope were used: (1) with a high-sensitivity and high-resolution charge-coupled device (CCD) camera, magnetic contrast imaging of the sample surface was performed with a spatial resolution of  $\sim 500$  nm; (2) the CCD camera was used as a conventional photodetector to measure the local magnetization reversal (hysteresis loops) by selecting an arbitrary region of interest (ROI) and thus a limited number of pixels. At maximum optical magnification ( $\times 100$  objective followed by a  $\times 4$  tube lens), each CCD pixel corresponded to an area of  $15 \times 15 \text{ nm}^2$  of the sample surface. Single-shot hysteresis loops of sample surface portions as small as  $1 \times 1 \mu\text{m}^2$  were measured<sup>49</sup>.

**BLS microscopy measurements.**  $\mu$ -BLS measurements were performed by focusing about 5 mW of monochromatic light (from a diode-pumped solid-state (DPSS) laser operating at a spectral line of 532 nm) with a microscope dark-field objective of large numerical aperture ( $\text{NA} = 0.75$ ) and super-long working distance (4.7 mm)<sup>50</sup>. Using the same objective, a coaxial viewing system based on a collimated light-emitting diode (LED) light source (455 nm wavelength), a beam expander and a CCD camera was implemented into the set-up to obtain a direct visualization of the laser spot and of the sample region under investigation. Electrical access to the sample was achieved using a specially designed ground-signal-ground (GSG) antenna with large extended pads where a d.c. was injected with a picoprobe.

During  $\mu$ -BLS measurements the sample holder and BLS spectrometer were controlled by TFPDAS4-MICRO and TFPDAS4 software, developed at Kaiserslautern University, which provided active stabilization of the lateral and vertical positions of the sample with a precision of  $\sim 50$  nm.

## References

- Nikulina, E., Idigoras, O., Vavassori, P., Chuvilin, A. & Berger, A. Magneto-optical magnetometry of individual 30 nm cobalt nanowires grown by electron beam induced deposition. *Appl. Phys. Lett.* **100**, 142401 (2012).
- Madami, M., Gubbiotti, G., Tacchi, S. & Carlotti, G. In *Solid State Physics* Vol. 63 (eds Camley, R. E. & Stamps, R. L.) 79–150 (Academic, 2012).



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