Supplementary information

Stretchable magneto-electronics

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(A) Photolithographically patterned GMR multilayers on PDMS

In order to fabricate GMR layer stacks on a free-standing rubber membrane, PDMS (Sylgard® 184) was first spin-coated onto silicon wafers. Optionally, an anti-stick photoresist layer was introduced to assist peeling the PDMS film from the rigid silicon support (Figure S1(a, b)). The PDMS precursor blend was cured in an oven at 90°C for 30 min under continuous nitrogen flow, resulting in a rubber film thickness of 40 μm (Figure S1(c)). GMR multilayer stacks consisting of Co(1 nm)/$[$Co(1 nm)/Cu(1.2 nm)$]_{50}$ were grown on the elastic PDMS surface (Figure S1(d)) using magnetron sputter deposition at room temperature (base pressure: 7.0 x 10^{-6} mbar; Ar sputter pressure: 7.5 x 10^{-4} mbar; deposition rate: 2 Å/s).

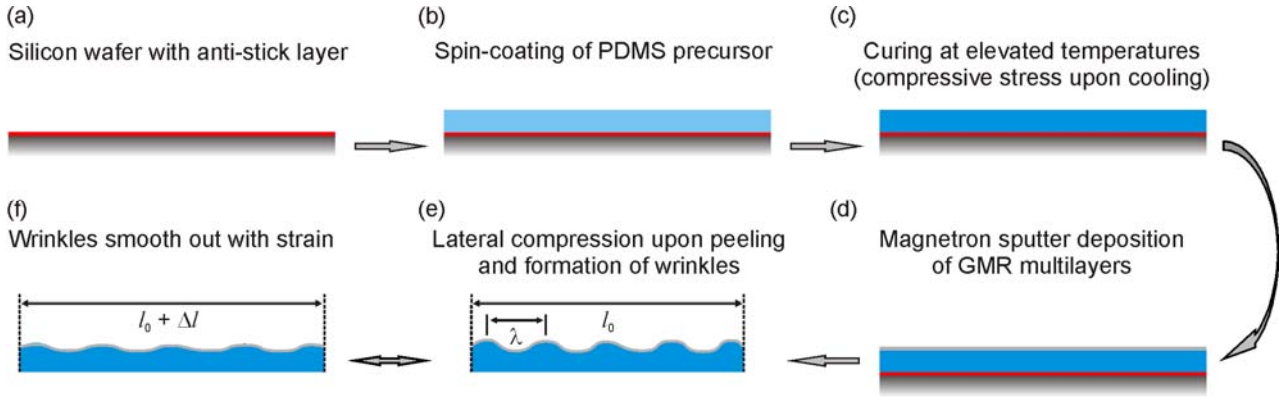

Figure S1 | Fabrication of the stretchable GMR multilayer stack: (a) SiOx wafer coated with photoresist as anti-stick layer. (b) The PDMS precursor blend is spin-coated on top. (c) The precursor is cured at elevated temperatures to form a rubber film. (d) Deposition of the GMR multilayers. (e) The metal coated rubber film is

peeled from the SiOx wafer. By peeling off the PDMS from the rigid support, wrinkling of the GMR multilayer stack takes place. (f) Upon elastic stretching the wrinkles are smoothened out and prevent the GMR layer from cracking, which allows for superior stretchability of the magnetic metal film.

The PDMS film was then peeled from the rigid silicon wafer leading to a free-standing elastic membrane covered with GMR multilayers (Figure S1(e)). In addition, photolithographic patterning on the PDMS surface before deposition of the metal films was performed to allow for electrical resistance measurements of the GMR films on the rubber substrates (Figure S2(a) and Figure 2(b) of the manuscript). This renders the fabrication process compatible to current microelectronic structuring procedures. The photolithographically structured GMR multilayers on top of a free-standing elastomeric PDMS membrane are shown in Figure 2(c) of the manuscript. After deposition, the rubber film with the patterned GMR stack was peeled from the SiOx wafer as shown in Figure S2(b). The peeling was assisted by an anti-stick layer and the peeling direction was along the patterned GMR structure. The resultant samples were free-standing PDMS rubber membranes coated with photopatterned $[Co/Cu]_{50}$ multilayers (Figure S2(c)).

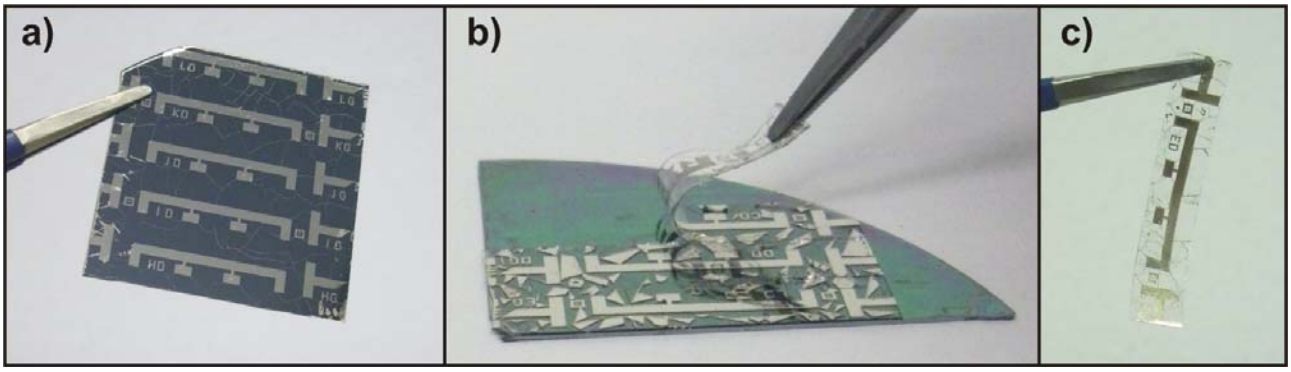

Figure S2: (a) Array of photopatterned $[Co/Cu]_{50}$ multilayers on a PDMS coated silicon wafer. (b) Peeling of the GMR multilayer coated PDMS rubber from the rigid silicon support by means of the anti-stick layer. (c) Photopatterned $[Co/Cu]_{50}$ multilayer on a free-standing PDMS membrane of 40 µm thickness. For the GMR films on free-standing rubber the formation of wrinkles is observed.

(B) AFM image of the GMR multilayer surface on top of a PDMS-coated SiOx wafer

After deposition of the $[Co/Cu]_{50}$ multilayer on top of a rubber coated silicon wafer, the roughness of the GMR film surface was measured before peeling by AFM (DI Dimension 3100). The rms-roughness value of the (5 x 5) µm² section shown in Figure S3 is 0.6 nm.

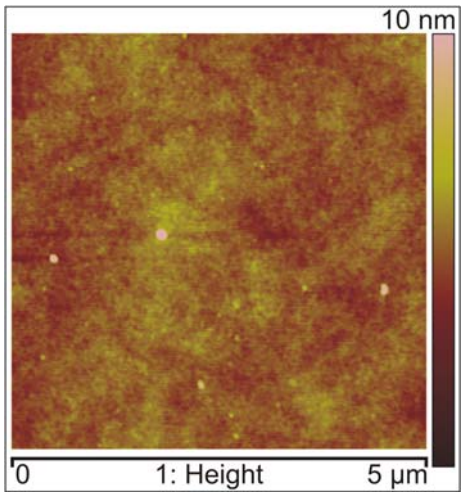

Figure S3: AFM micrograph of the GMR multilayer surface on a PDMS coated silicon wafer. For the shown surface section of (5 x 5) µm² the value of the root mean squared roughness is 0.6 nm.

(C) Wrinkling of the [Co/Cu] magnetic film on a PDMS membrane

On the free-standing rubber membrane, the formation of wrinkles is observed (Figure 2(c) of the manuscript). This buckling of the metal film occurs due to a contraction of the underlying PDMS upon peeling. The thermal shrinkage of the rubber film upon cooling down after curing is suppressed by the rigid silicon wafer (Figure S1, process step (c)). This suppression is due to a large mismatch of the thermal expansion coefficients of the two materials (9.6×10^{-4} K^{-1} (PDMS [PDMS Data Sheet]) vs. 2.6×10^{-6} K^{-1} (silicon [Lyon 1977])). As a result, the thermal contraction of the rubber is "stored" by means of a compressive lateral stress arising inside the PDMS. This stress is maintained during the sputter deposition of the GMR layers and is not released until the sample is peeled from the rigid supporting wafer. Upon peeling, the rubber contracts which causes wrinkling of the incompressible metal film. The formation of wrinkles after film deposition onto pre-strained elastic substrates has been exploited previously for compliant electrodes, where stretchability is provided by smoothing out the buckles for lateral strains perpendicular to them [Lacour 2003] (Figure S1(f)). The thermal contraction of the PDMS for the temperature difference of $\Delta T = 70°C$ (from 90°C down to room temperature), is about 7%. Figure S4(a) shows a confocal microscope (SDCM) image of the wrinkled GMR film.

The height profile of the sample reveals a wrinkling period of about 17 µm and a mean amplitude of about 0.5 µm (Figure 3(a) of the manuscript). A focussed ion beam (FIB) cut of the sample (SEM inset in Figure 3(b) of the manuscript) discloses the wavy GMR film (indicated by grey line) firmly attached to the bulky rubber (highlighted in blue). This suggests that the contact between the PDMS and the metal film is maintained throughout the wrinkle structure. As a result, we can apply a theoretical model by Bowden et al. [Bowden 1998] to estimate the period of the wrinkles, λ, formed for a thin incompressible metal film of thickness *t* on an elastomeric surface:

$$\lambda = 4.36\, t \left(\frac{E_f \left(1 - \nu_p^2\right)}{E_p \left(1 - \nu_f^2\right)} \right)^{1/3} \qquad (1)$$

here $E_{p,f}$ and $\nu_{p,f}$ denote Young's module and the Poisson's ratio of the metal film (f) and the polymer (p), respectively. The values of these parameters used for the simulations are as follows: E_p = 1.6 MPa, E_f = 171 GPa ($E_{Co} \approx 211$ GPa, $E_{Cu} \approx 131$ GPa), ν_p = 0.48, ν_f = 0.33 (ν_{Co} = 0.31, ν_{Cu} = 0.35). The Young's modulus for the PDMS was determined from analyzing stress-strain measurements (see Supplementary Information, part F), while the Poisson's ratio and the data for the metal film is taken from the literature [Bowden 1998, Sanders 1997, Doi 1970]. Considering the total thickness of the GMR multilayer stack ($t \approx 110$ nm), the calculation according to Eq. (1) predicts a value for the wrinkling wavelength of $\lambda \approx 22$ µm, which is in good agreement with the value derived from the line scan in Figure 3(a) of the manuscript.

A set of optical micrographs of the wrinkled GMR film at different uniaxial strains is shown in panels (I-III) in Figure S4(b). With increasing strain, wrinkles become less pronounced indicating that the metal film becomes smoother. Cracks emerging in the metal film upon stretching are not observed.

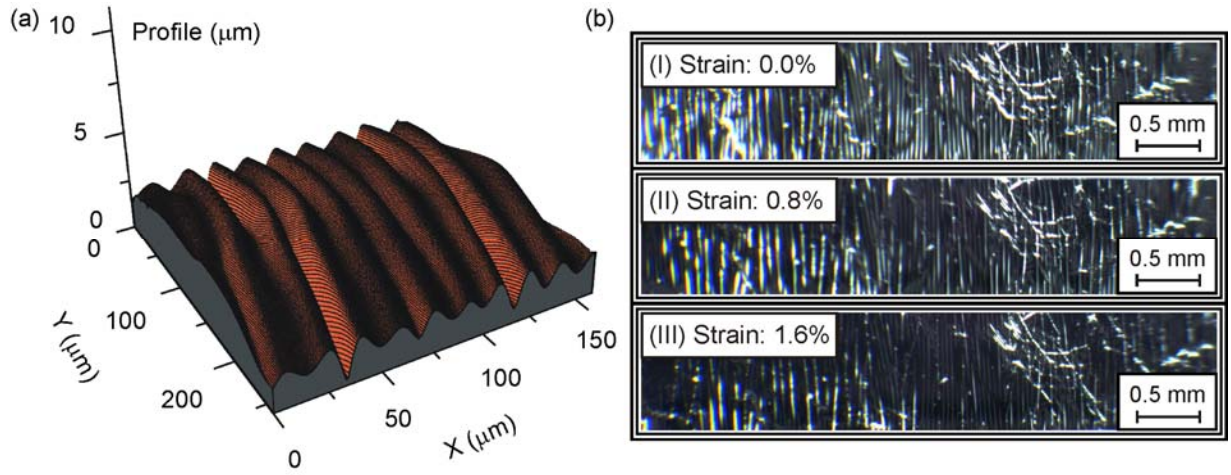

Figure S4: Thermally induced wrinkling of GMR multilayers on top of a free-standing rubber membrane. (a) Confocal microscopy image of the sample surface. (b) Optical microscopy images. Panels (I-III) reveal the modification of the samples morphology upon stretching. For larger strain values, wrinkles become less pronounced indicating that metal film smoothens out.

(D) Stretching of wrinkled GMR multilayers on free-standing rubber membranes

In the applied method, a bar of PDMS (3 mm x 5 mm x 50 mm) was fixed between two clamps and pre-stretched to a length of 60 mm (20%). The actual GMR multilayers on the 40-µm-thick PDMS membrane were then mounted on top of this elastic holding bar (Figure S5(a)).

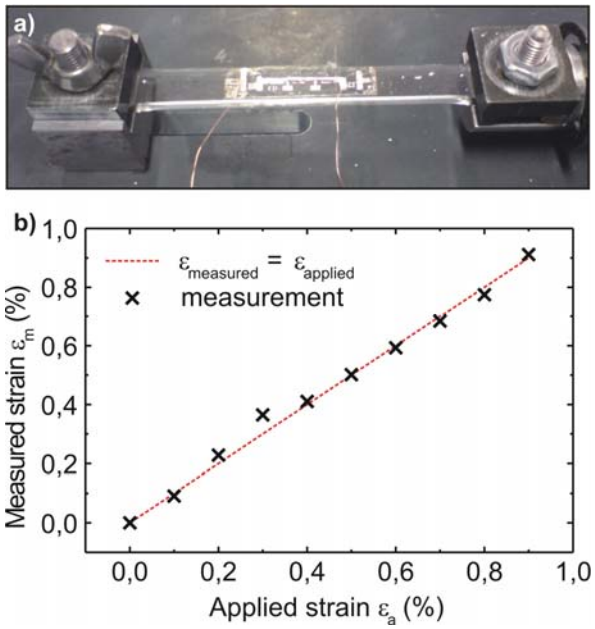

Figure S5: (a) Piggyback setup for application of tensile strain to the GMR multilayers on rubber substrates. A bar of PDMS (3 mm x 5 mm x 50 mm) is applied between two clamps, one of which can be accurately moved to apply a tensile strain along the bar. The bar is pre-stretched to a length of 60 mm (20%) to avoid slack. On top of the pre-stretched holding bar a sample of photolithographically patterned GMR multilayer on free-standing PDMS membrane is applied. For the measurement of the electrical resistance two thin copper wires (0.1 mm) are glued to the GMR multilayer with silver paste. (b) The strain measured directly on the GMR multilayer with an optical microscope is in agreement with the strain applied to the PDMS holding bar.

The two smooth PDMS surfaces provide a good adhesion of the sample to the holding bar. This method ensures on the one hand that there is minimal strain on the sample at the beginning of the measurement and on the other hand that there is no slack also and the sample is stretched as soon as the separation of the clamps is increased. Thin copper wires (0.1 mm) were connected to the two ends of the GMR multilayer structure with silver paste to measure the electrical resistance while stretching. The strain applied on the GMR multilayer was checked with an optical microscope. As shown in Figure S5(b) the strain measured on the multilayer is in good agreement to the strain applied to holding bar, proving the good adhesion to the PDMS membrane.

(E) Detection of a rotating magnetic field with elastic GMR sensor on a curved surface

For this experiment, a GMR sensor element on PDMS membrane was attached to a plastic foil shaped in a ring geometry (Figure S6). A permanent magnet was rotating and the change of the resistance of the GMR sensor was recorded versus time. When the magnet is in proximity to the GMR sensor, a clear decrease of the samples resistance was detected (Supplementary video 1). The performance of the GMR sensor was cross-checked using a conventional Hall-effect sensor.

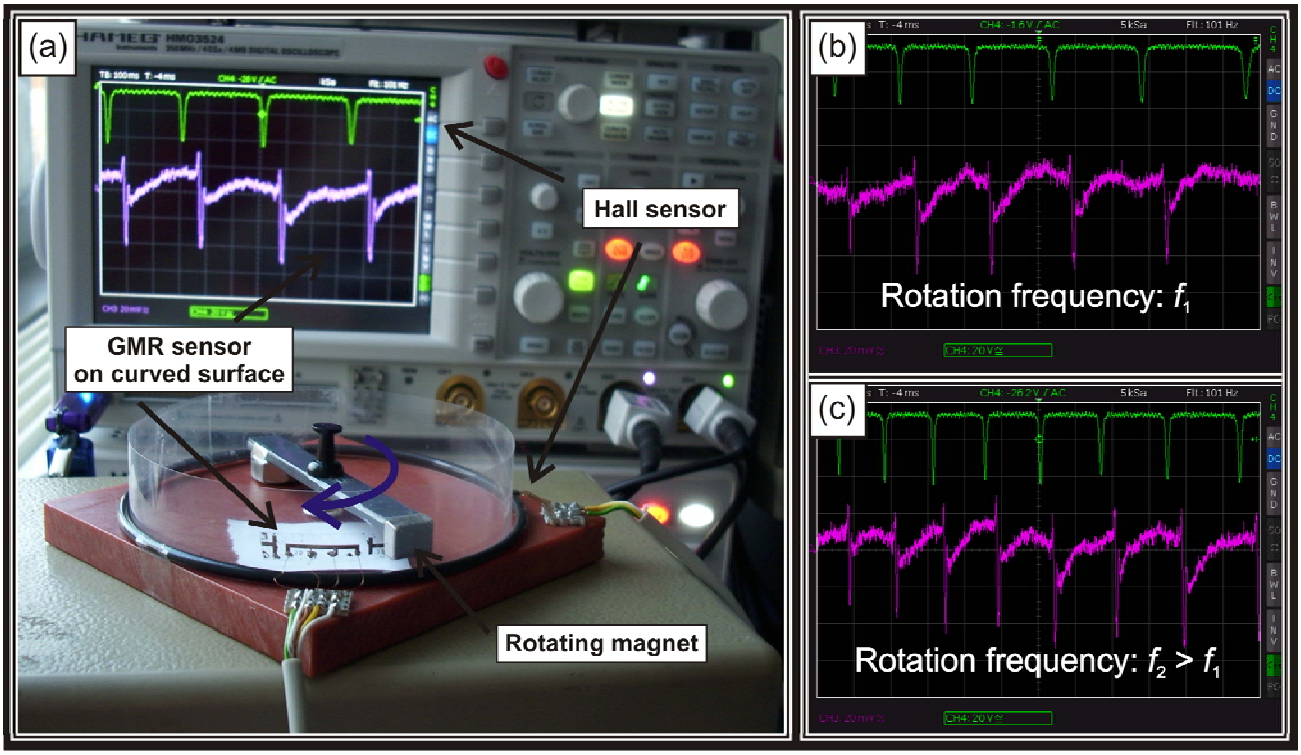

Figure S6: (a) Experimental set-up used for measurement of the rotating magnetic field of a permanent magnet with the GMR sensor on an elastic PDMS membrane attached to the curved surface of a plastic foil. When the magnet approaches the GMR sensor, decrease of the sample resistance is measured revealed by the spikes in an oscilloscope signal (purple curve). The green curve represents the reference signal taken using a conventional Hall-effect sensor. The signal from the GMR sensor and the Hall-effect sensor are phase-shifted due to the different spatial position of both sensor elements. (b, c) Response of the GMR sensor on the rotating magnetic field of different frequency: The increase of the rotation frequency of the permanent magnet can be easily traced with the GMR sensor.

(F) Mechanical characterization of the elastic PDMS membrane

PDMS membranes were prepared for mechanical characterization measurements in a stress-strain setup. The specimens were 15 mm wide and the film thickness was 40 µm as for the membranes coated with GMR multilayers. The initial length between the clamps was 40 mm. The figure below shows the stress-strain behavior of a rubber membrane stretched to about 50% and back at a constant rate of 2 mm/min. The black and gray curves (Figure S7) represent the stress upon loading and unloading, respectively. The two curves are in good agreement, which justifies the strong elastic behavior of the PDMS material. The Young's modulus of the rubber is derived from the slope of the loading curve at the first 5% strain, which is indicated by the dotted red line. The obtained value is E_{PDMS} = 1.6 MPa.

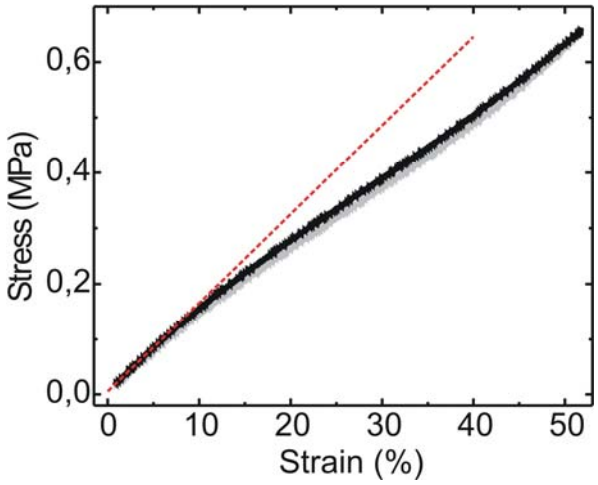

Figure S7: Stress-strain curve of a PDMS film (40 mm x 15 mm x 40 µm) showing rubber-like behavior. Black curve is for loading, gray curve is for unloading. For the determination of the Young's modulus the linear fit taking into account the first 5% of strain is shown in red (E_{PDMS} = 1.6 MPa).

Supplementary video 1: Performance of the GMR sensor on an elastic PDMS membrane attached to the curved surface of the plastic foil.

References

[Bowden 1998] N. Bowden et al., *Nature* **393**, 146 (1998).

[Doi 1970] H. Doi et al., *Metallurgical Transactions* **1**, 1417 (1970).

[Lacour 2003] S.P. Lacour et al., A*pplied Physics Letters* **82**, 2404 (2003).

[Lyon 1977] K.G. Lyon et al., *Journal of Applied Physics* **48**, 865 (1977).

[PDMS Data Sheet] Dow Corning, Sylgard® 184 technical data sheet.

[Sanders 1997] P.G. Sanders et al., *Acta Materialia* **45**, 4019 (1997).