Supporting Information

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Local Photothermal Control of Phase Transitions for On-Demand Room-Temperature Rewritable Magnetic Patterning

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Supporting Information for “Local Photothermal Control of Phase Transitions for On-Demand Rewritable Magnetic Patterning”

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S1. TEMPERATURE-DEPENDENT IMAGING OF Fe$_{0.52}$Rh$_{0.48}$

In this section we image the global magnetic domain structure of the device in Fig. 3e as a function of temperature, with no local FM writing involved. All subsequent ANE images – unless specified otherwise – are half-differences between $H_{app} = \pm 1$ kOe to isolate $V_{ANE}$ from non-magnetic artifacts and are acquired at $f = 0.6$ mJ/cm$^2$ laser fluence.

Fig. S1: Temperature-dependent imaging of Fe$_{0.52}$Rh$_{0.48}$. Anomalous Nernst images of the sample in Fig. 3f of the main text at 295 K in the AF phase, within the transition region at 350 K, in the uniform FM phase at 390 K, and at 295 K after cooling in zero magnetic field. Weak contrast at 295 K in the AF phase arises from unpinned uncompensated moments. Uniform $V_{ANE}$ at 390 K arises from a uniform FM phase. At 295 K after cooling, submicron FM domains interspersed with AF domains indicate coexisting AF and FM phases.

Fig. S1 shows ANE images at 295 K in the AF phase, at 350 K within the transition region, at 390 K in the FM phase, and at 295 K after cooling in zero magnetic field. At 390 K, $V_{ANE}$ is spatially uniform aside from highlighted scratches on the sample surface, which is consistent with previous ANE imaging of ferromagnets [1–3] and indicates a uniform FM phase. Interestingly, $V_{ANE}$ is nearly spatially uniform even at 350 K although its magnitude is weaker than at 390 K. This contrast may be due to pinned uncompensated moments in the AF phase becoming unpinned before transitioning to the FM phase, as found in our previous FeRh work[4]. At 295 K after cooling, smaller FM domains interspersed with AF domains indicate coexisting AF and FM phases. This result is consistent with the $M(T)$ measurement in Fig. 2 of the main text, in which $M(295 \text{ K})$ after cooling from the FM phase is intermediate between the values of $M(295 \text{ K})$ in the AF phase and $M$ in the FM phase. It
FIG. S2: Imaging a written FM region as a function of magnetic field. a, Imaging a 5×5 μm written FM square as a function of applied field $H_{app}$, from $|H_{app}| = 100$ Oe to 1 kOe. Most written FM domains switch between $|H_{app}| = 200$ Oe and $H_{app} = 600$ Oe, while the unpinned uncompensated moments in the surrounding AF regions switch at $|H_{app}|$ between 600 Oe and 1 kOe. b, Half-difference image of the entire device between $H_{app} = \pm 3$ kOe. At room temperature, written FM domains remain stable up to the largest magnetic fields we can apply (3 kOe).

also demonstrates that the spatial nonuniformity of laser-written FM regions is an intrinsic property of the material film and not an artifact of laser heating.

S2. EFFECTS OF MAGNETIC FIELD ON WRITTEN FM REGIONS

In Fig. S2, we image a 5 μm ×5 μm written FM square within a 10 × 24 μm device as a function of magnetic field $H_{app}$ at 295 K. Fig. S2a shows half-differences between positive and negative $H_{app}$ as a function of $|H_{app}|$ from 100 Oe to 1 kOe. Due to exchange coupling between coexisting AF and FM phases, the switching field of the written FM regions is spatially inhomogeneous and in general greater than the $\approx 100$ Oe switching field in the uniform FM phase. Written FM domains begin to switch at 200 Oe and most have switched at 400 Oe, although a few continue to rotate up to $|H_{app}| = 1$ kOe. The unpinned uncompensated moments in the surrounding AF phase are more strongly coupled to the bulk Néel order than the written FM regions are and therefore have higher saturation field - between 600 Oe and 1 kOe.

Fig. S2b shows the half-difference at $|H_{app}| = 3$ kOe, the largest field we can apply in situ. The structure of both the written FM region and the surrounding uncompensated moments
is almost identical to the structure at $|H_{\text{app}}| = 1$ kOe in Fig. S2a. Therefore, at least up to $H_{\text{app}} = 3$ kOe, FM domains rotate, but do not move or grow - their position is stable under magnetic field. This result shows that taking the half-difference between $\pm 1$ kOe completely separates magnetic $V_{\text{ANE}}$ signal from non-magnetic charge Seebeck artifacts. In addition, magnetic field does not affect the background electrical noise level, and therefore does not affect the signal-to-noise once the moments are saturated.

**S3. EFFECTS OF ELEVATED GLOBAL TEMPERATURE ON WRITTEN FM REGIONS**

In Fig. S3 we image the same written 5 $\mu$m $\times$ 5 $\mu$m FM square as in Sec. S2 as a function of temperature. Here we show half-differences between $H_{\text{app}} = \pm 1$ kOe; note that although a magnetic field decreases the transition temperature in FeRh, $H_{\text{app}} = 1$ kOe lowers $T_C$ by $< 1$ K [4]; therefore its effects are negligible.

![Image](FIG. S3: Imaging written FM domains at elevated temperature. The same written FM region as in Fig. S2 is imaged at and above room temperature. Although FM domains are not significantly affected at elevated temperature, pinned moments in the surrounding AF regions become unpinned before transitioning into the FM phase. The maximum bath temperature at which the FM region is distinguishable from the AF background is 318 K, 23 K above ambient temperature.

Written FM regions are largely unaffected by elevated temperature; however, the contrast between written FM regions and the surrounding AF background weakens as pinned moments become unpinned and eventually transition into the FM phase. The maximum temperature at which written FM regions can be distinguished from the AF background is
318 K, 23 K above ambient temperature.

S4. RETENTION TIME OF FM REGIONS AT ROOM TEMPERATURE

![Image of FM regions comparison](image)

FIG. S4: Imaging the 5×5 µm FM square after initially writing and after 3 days at room temperature.

Almost no change in domain structure shows that the FM regions are stable for at least 3 days at ambient.

In this section we investigate the retention time of FM regions; in other words, how long written FM regions remain stable at room temperature. The experiments in Secs. S2 and S3 were taken over a period of several days using the same 5×5 µm written FM square. In Figure S4 we replot half-difference ANE images from Figs. S2 and S3, taken 3 days apart, together with the difference between these half-differences. The difference shows very slight changes to the FM domain structure, which may be due to imperfect image alignment between four anomalous Nernst images. Therefore, written FM regions are stable for at least several days.

S5. TEMPERATURE PROFILE FROM LASER HEATING

A. Laser-induced temperature profile as a function of laser fluence

In this section we determine the spatiotemporal thermal profile from laser heating as a function of laser fluence, using a combination of finite-element simulations and measurements of local heating-induced changes in resistivity that is detailed further in our previous work[1]. The thermal profile is written in cylindrical coordinates as $\Delta T(r, z, t)$, where $r$ is the radial distance from the beam center and $z$ is the depth defined relative to the film surface.

$\Delta T(r, z, t)$ is simulated by numerically solving the radially symmetric heat equation using the COMSOL Multiphysics® software package. The laser is modeled as a distributed heat source that exponentially decays with $z$ according to the skin depth of FeRh. Because the
FIG. S5: **Laser-induced temperature profile in FeRh**, determined using a combination of finite-element simulations and measurements of local laser-induced changes of resistivity. **a**, Temperature change $\Delta T(t)$ at the beam center ($r = 0$) averaged over film depth ($z$), at the $f = 8.0 \text{ mJ/cm}^2$ fluence used for writing. The maximum $\Delta T_{\text{max}} \approx 90 \text{ K}$ occurs at 20 ps after pulse arrival, after which $\Delta T(t)$ decays approximately exponentially. **b**, Temperature depth profile $\Delta T(z)$ at $r = 0$ at three times after pulse arrival. **a** and **b** are results from finite-element simulations, scaled to match the laser-induced resistivity measurements in **c**. $\Delta T(t)$ causes a transient resistivity increase $\Delta \rho(t)$, which produces a voltage $V(t)$ when a current $I_{\text{DC}}$ is applied to the device. After amplification and mixing, $V(t)$ becomes a lock-in voltage $V_{\text{LI}}(I_{\text{DC}})$ with constant slope $dV_{\text{LI}}/dI_{\text{DC}}$. **c**, Measured slope $dV_{\text{LI}}/dI_{\text{DC}}$ as a function of laser fluence, used to correctly scale the simulations. At the reading ($0.8 \text{ mJ/cm}^2$) and writing ($10.8 \text{ mJ/cm}^2$) fluences, $\Delta T_{\text{max}} = 6.5 \text{ K}$ and $90 \text{ K}$, respectively.

Skin depth in Fe$_{0.52}$Rh$_{0.48}$ (42 nm, from Ref.[5]) is comparable to the 35 nm film thickness, a complicated series of internal reflections occurs and as a result the overall fraction of light absorbed is not well-known. We account for this uncertainty by assuming the simulated $\Delta T(r, z, t)$ is accurate up to an overall scaling coefficient $\gamma$, which must be experimentally determined. Correctly scaled simulation results are presented first, followed by the procedure for determining $\gamma$.

Fig. S5a shows the depth-averaged temperature evolution at the beam center ($r = 0$) as a function of time after pulsing at $t = 0$ at the writing fluence - 10.7 mJ/cm$^2$. The maximum averaged $\Delta T$ is $\sim 90 \text{ K}$, occurring 20 ps after the laser pulse arrival, after which $\Delta T$ decays approximately exponentially in time. Note that because the electron-phonon thermalization time is $<1 \text{ ps}$ in metals[6] and the laser pulse is $\sim 3 \text{ ps}$ long, the electrons and phonons are in thermal equilibrium throughout laser heating and a one-temperature model is sufficient. Fig. S5b presents $\Delta T(r = 0)$ as a function of $z$ at three different times after heating. After 100 ps, the sample thickness is nearly uniform in temperature.
We now describe an abbreviated procedure for experimentally determining the scaling factor \( \gamma \); further details can be found in Ref.[1]. \( \Delta T(r, z, t) \) causes a transient local increase in the FeRh resistivity \( \Delta \rho (r, z, t) \). When a DC current \( I_{DC} \) is applied to a patterned device, \( \Delta \rho (r, z, t) \) produces a transient voltage drop \( V(t) \) across the device contacts proportional to the volume integral of \( \Delta \rho (r, z, t) \). After amplification and homodyne mixing, \( V(t) \) is transduced into a measurable lock-in voltage \( V_{LI}(I_{DC}) \). In the linear regime where \( \Delta \rho = \alpha \Delta T \) with constant temperature coefficient of resistivity \( \alpha \), the lock-in slope \( dV_{LI}/dI_{DC} \propto \Delta T(r, z, t) \). Given \( \alpha \) – measured to be \( 2.9 \times 10^{-3} \text{ K}^{-1} \) in both the AF and FM phases – \( dV_{LI}/dI_{DC} \) can be calculated from the simulated \( \Delta T(r, z, t) \). \( \gamma \) is determined by scaling \( \Delta T(r, z, t) \) such that the simulated and measured \( dV_{LI}/dI_{DC} \) are equal.

Fig.S5(c) plots the measured \( dV_{LI}/dI_{DC} \) in the device from Fig. 3f as a function of fluence \( f \). Note that although \( dV_{LI}/dI_{DC} \) is linear in \( f \) in the AF (\( f < 2 \text{ mJ/cm}^2 \)) and FM (\( f >= 6 \text{ mJ/cm}^2 \)) phases with the same slope, it is sublinear in \( f \) within the transition region as larger fractions of material convert to the lower-resistance FM phase. In the calculation \( \Delta T(r, z, t) \) is assumed to be linear in \( f \), which is true in the AF and FM phases, but may not be strictly true in the transition region. Nevertheless, \( \Delta T \) obtained for FM regions written at intermediate fluences in Fig. S5 is consistent with the bulk \( M(T) \) measurements in Fig. 2a. We determine the peak temperature increase \( \Delta T_{max} \) to be 6.5 K at the imaging fluence (0.8 mJ/cm\(^2\)) and 90 K at the writing fluence (10.7 mJ/cm\(^2\)).

**B. Fluence dependence of FM writing**

To confirm our temperature calibration and determine the minimum fluence required to induce the magnetic phase transition, we image sample regions irradiated with increasing laser fluence between \( f = 3.6 \text{ mJ/cm}^2 \) and 10.7 mJ/cm\(^2\). The images are acquired directly in succession, rather than resetting the AF state in between changing the writing fluence value. The resulting half-difference ANE images, along with the calibrated peak temperature rises \( \Delta T_{max} \), are shown in Fig. S6; an image of the unirradiated AF area is also provided for reference. Pulsing with \( f = 3.6 \text{ mJ/cm}^2 \) (\( \Delta T_{max} = 31 \text{ K} \)) causes minimal changes in the ANE contrast, which indicates that 31 K is insufficient to locally induce the FM phase transition of Fe\(_{0.52}\)Rh\(_{0.48}\). In distinct contrast, when fluence values are increased to between 6.3 and 10.8 mJ/cm\(^2\) (\( \Delta T_{max} = 56 \text{ K} - 90 \text{ K} \)), clear changes associated with the FM phase transition
FIG. S6: Laser fluence dependence of FM writing. a, The digital mask used to test fluence dependence of writing: a single pixel, squares of 1, 2, 3, and 4 μm dimension, and 1×4 μm and 4×1 μm rectangles. b, Images before writing and after writing the pattern in (a) at increasing fluence. Images are labeled by the writing fluence employed, the maximum local change in temperature ΔTmax, and the local maximum temperature Tmax.

are detected in the ANE images. Above f = 8.7 mJ/cm² (ΔTmax = 77 K), the ANE contrast changes begin to saturate as most irradiated AF areas have transitioned entirely to the FM phase. These findings are consistent with the macroscopic film transport and magnetisation measurements presented in Fig. 2 of the main text.

S6. NOTE ON REWRITABILITY AND ERASING OF Fe₀.₅₂Rh₀.₄₈

All writing and erasing in Fig. 3f, Fig. 4, and the supporting information was performed on a single 10 μm × 24 μm device of Fe₀.₅₂Rh₀.₄₈, during which the device was written and erased a total of 8 times. No irreversible changes in ANE contrast that would indicate surface damage from laser ablation were measured. Device resistance is another measure of sample damage – it first decreases as the film is annealed by laser heating and then increases as ablation occurs. After every reset the device recovered its original resistance of 80.6 Ω ± 0.3 Ω (the large uncertainty is due to variations in contact resistance from repeated wire bonding). We therefore conclude within experimental uncertainty that ΔTmax = 90 K from laser heating and thermal cycling by repeatedly cooling with liquid nitrogen and
warming to room temperature do not adversely affect the FeRh film quality.

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