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Magnetic phase dependency of the thermal conductivity of FeRh from thermorefectance experiments and numerical simulations

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FeRh is well known in its bulk form for a temperature-driven antiferromagnetic (AF) to ferromagnetic (FM) transition near room temperature. It has aroused renewed interest in thin film form, with particular focus on its biaxial AF magnetic anisotropy which could serve for data encoding, and the possibility to investigate laser-assisted phase transitions, with varying contributions from electrons, phonons and magnons. In order to estimate the typical temperature increase occurring in these experiments, we performed modulated thermorefectance microscopy to determine the thermal conductivity κ of FeRh. As often occurs upon alloying, and despite the good crystallinity of the layer, κ was found to be lower than the thermal conductivities of its constituting elements. More unexpectedly given the electrically more conducting nature of the FM phase, it turned out to be three times lower in the FM phase compared to the AF phase. This trend was confirmed by examining the temporal decay of incoherent phonons generated by a pulsed laser in both phases. To elucidate these results, first and second principles simulations were performed to estimate the phonon, magnon and electron contributions to the thermal conductivity. They were found to be of the same order of magnitude, and to give a quantitative rendering of the experimentally observed κ_{AF} . In the FM phase however, simulations overestimate the low experimental values, implying very different (shorter) electron and magnon lifetimes.

INTRODUCTION

The FeRh alloy near equiatomic composition is a unique magnetic material, studied long ago in bulk form^{1,2}, and which has recently sparked a renewed interest in thin film form for technological applications. The first-order metamagnetic phase transition of FeRh from the antiferromagnetic (AF) to the ferromagnetic (FM) state happens close to room temperature, and the associated changes in magnetic, electrical and thermodynamical properties have been utilized to propose novel approaches for memory cells³, heat-assisted magnetic recording⁴, and magnetic refrigeration⁵. FeRh thin films are also good candidates to investigate laser-assisted transient⁶⁻¹² or irreversible¹³ phase transitions.

Excitation of the AF phase by a femtosecond laser pulse generates an out-of-equilibrium population of electrons and then phonons, whose temperatures eventually equilibrate, typically on picosecond time scales. The respective roles of electrons and phonons at the onset of a transient FM state are still much debated^{7,8,10,11,14}. After this initial step on the ps timescale the local temperature decreases through heat diffusion. This evolution determines the nucleation, expansion and coalescence of FM domains as well as the decay of the transient magnetic moment upon cooling back to the AF phase¹². However, the heat diffusion coefficient D of FeRh is poorly known. It is related to the thermal conductivity κ by $D = \kappa/(\rho C)$ where ρ is the mass density and C the specific heat. Both ρ and C are known to vary with temperature and magnetic phase^{15,16}. The heat conductivity also determines the stationary temperature rise of the sample when the pulsed laser repetition rate is too high for the system to relax

back to its base temperature between pulses^{6,8,17-19}, or when a CW laser or current is used to induce a local nucleation of FM domains^{13,20}. Whereas specific heat measurements are reported for FeRh bulk^{1,2,21} and thin film samples¹⁶, the thermal conductivity is usually estimated from the electrical conductivity σ using the Wiedemann-Franz law $\kappa_e = L_0 T \sigma$, with L_0 being the Lorenz number⁷. A noteworthy exception is a recent paper by Ahn *et al.*¹⁹ in which they evaluate indirectly $\kappa_{AF}=11.8$ and $\kappa_{FM}=22.4$ W m⁻¹ K⁻¹.

There are numerous reports of the material's electronic and phononic band structures, some expliciting calculations of its thermal properties²²⁻²⁷. Since FeRh is metallic, one could expect the electronic contribution to the thermal conductivity κ_e to be dominant over the phononic and magnonic (κ_{ph} , κ_m) ones, and to govern how κ should scale between its two magnetic phases. Relating in first approximation κ_e to the electrical conductivity using the Wiedemann-Franz law mentioned above gives a value almost twice larger in the FM phase as in the AF phase²⁸. As for the phononic contribution, recent DFT calculations by Cazorla *et al.*²⁶ predict it to be very slightly smaller in the AF than the FM phase. Finally, there is no record yet – either experimental or numerical – for the magnonic contribution κ_m . In pure BCC Fe it has been estimated to represent about 10% of the total thermal conductivity²⁹. Overall, calculations so far suggest the FeRh FM phase to be thermally more conductive than the AF one.

In this paper we determine the total thermal conductivity of a 195 nm FeRh thin film, using modulated thermorefectance (TR) microscopy. It is an ideal technique to measure the thermal properties of thin films deposited on a substrate, when

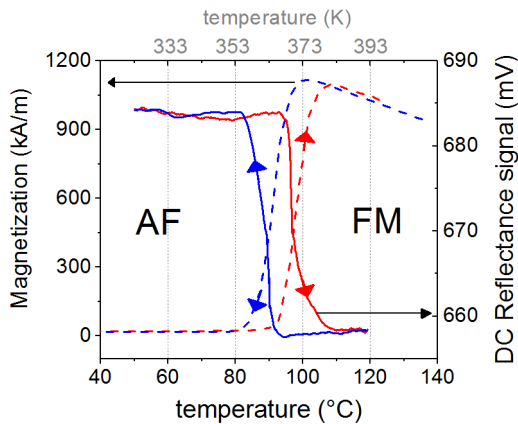


FIG. 1. Determination of the transition temperature of the film. Left: Global measurement of magnetization using vibrating sample magnetometry (dashed line). Right: Local measurement of the DC reflectance (continuous line) under probe beam only ($\lambda = 488$ nm, $0.8 \mu\text{m}$ spatial resolution) with a temperature ramp of 2 K/min .

the volume is too small to be probed by standard calorimetry techniques^{30–33}. This enables us to estimate quantitatively transient and stationary laser-driven temperature rises. We confirm the results on thermal conductivity by monitoring the time decay of interferometric measurements in a pump-probe setup in both AF and FM phases. In parallel, we compute the electronic, magnetic and vibrational properties of the material, and the different contributions to the thermal conductivity in both phases. We then discuss how the total value compares to our data.

SAMPLE

An FeRh film with a thickness of $h = 195$ nm was deposited on a $500\text{-}\mu\text{m}$ -thick MgO(001) substrate via magnetron sputtering of an equiatomic FeRh target. The film was grown at 430°C after preheating the substrate for 60 min at the same temperature. We used an Ar pressure of 2.7×10^{-3} mbar and a sputtering power of 50 W, leading to a deposition rate of 2 nm min^{-1} . Post-growth annealing in high vacuum at 780°C for 80 min yielded a good quality and homogeneous CsCl-type structure of the film¹⁵. It is nearly fully relaxed, with a $+0.03\%$ strain in the out-of-plane direction at room temperature. Finally, a 2-nm-thick Pt cap was grown after cooling the sample below 120°C , to protect the film from oxidation.

The temperature-dependent magnetization data of the film measured via vibrating sample magnetometry (VSM) (Fig. 1) show a heating transition from AF to FM state between 87 and 108°C (360 and 381 K), and a cooling transition between 100 and 80°C (373 and 353 K). The very weak residual ferromagnetic contribution in the AF phase (4% of the magnetization at 400 K) is a signature of the excellent quality of the sample. The AF-FM transition can also be monitored locally by reflectance microscopy at variable temperature. For this the sample is glued with thermal paste on the holder of a Linkam cryostat adapted to a microscopy set-up. As ex-

pected for FeRh^{34–36}, a 3–4% hysteretic change of the phase-dependent reflectance $R(T)$ is observed at the AF-FM transition (Fig. 1). The slight difference in shape and position of the transition compared to the curve measured by VSM reflects the much smaller probed volume (a few μm^2 wide, and about 10 nm deep) and the spatial variations of the magnetic properties.

MODULATED THERMOREFLECTANCE EXPERIMENTS: METHODOLOGY AND RESULTS

We now briefly describe the home-built thermorefectance microscope that allows to access the thermal properties of the sample.

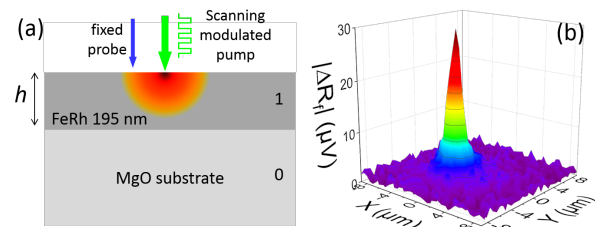


FIG. 2. (a) Schematics of the modulated thermorefectance set-up. (b) $10 \times 10 \mu\text{m}^2$ map of the amplitude of the modulated thermorefectance signal.

Light from a $\lambda = 488$ nm (blue) continuous wave (CW) laser (probe beam) is focused onto its surface to a diffraction-limited spot by a $\times 50$ long-working-distance objective (numerical aperture $\text{NA} = 0.5$, working distance 10.6 mm). The typical power impinging on the sample is $120 \mu\text{W}$. A heat source is provided by a 532 nm (green) CW laser passing through an acousto-optic modulator driven by a square modulation at frequency f and focused onto the sample by the objective (≈ 5.3 mW on the sample). This green laser spot is raster-scanned on the sample surface while the reflected blue probe beam is collected by the photodiode which monitors the reflectance of the layer. The green light reflected back towards the detector is blocked by an interference filter. The signal is fed into a lock-in amplifier that returns the amplitude and phase of the AC component of the reflectance at the frequency of the modulated heat source, ΔR_f , as well as the DC component R_0 thanks to an Analog to Digital Converter (ADC). A typical map of the modulated reflectance is shown in Fig. 2(b) (amplitude component).

As described in detail in Ref. 30, the reflectance is assumed to depend on the temperature increase to the first order $R(\mathbf{r}, t) = R_0 + \frac{\partial R_{probe}}{\partial T} I_{probe}(\mathbf{r}) * \Delta T(\mathbf{r}, t)$, where the temperature profile is convolved with the probe spot profile $I_{probe}(\mathbf{r})$. The modulated reflectance ΔR_f at frequency f reflects the f -harmonic of the temperature increase ΔT_f induced by the pump laser. Modulation frequencies are moreover assumed to be low enough to consider the system at thermodynamic equilibrium, with the thermal flux being proportional to the tem-

161 perature gradient³⁰. Adapting Ref. 37 and using cylindrical
162 coordinates, the thermal field at the surface of the sample and

163 the modulated reflectance are given by inverse Hankel trans-
164 forms as detailed in Eqs. (1-3).

$$\Delta T_f(r) = \frac{Q_f}{2\pi} \int_0^\infty g(u) J_0(ur) u \exp\left(-\frac{d_{\text{pump}}^2 u^2}{32}\right) du, \quad (1)$$

$$\Delta R_f(r) \propto \frac{\partial R_{\text{probe}}}{\partial T} \int_0^\infty g(u) J_0(ur) u A(u) \exp\left(-\frac{d_{\text{pump}}^2 u^2}{32}\right) du, \quad (2)$$

$$g(u) = \frac{\alpha^2}{\alpha^2 - \Sigma_1^2} \frac{\left(1 - \frac{\kappa_0^* \Sigma_0}{\kappa_1 \alpha}\right) (\cosh(\Sigma_1 h) - \exp(-\alpha h)) + \left(\frac{\kappa_0^* \Sigma_0}{\kappa_1 \Sigma_1} - \frac{\Sigma_1}{\alpha}\right) \sinh(\Sigma_1 h)}{\kappa_0^* \Sigma_0 \cosh(\Sigma_1 h) + \kappa_1 \Sigma_1 \sinh(\Sigma_1 h)}, \quad (3)$$

165 where $\Sigma_i(u, f) = \sqrt{u^2 + j \frac{2\pi f}{D_i}}$ is akin to a complex thermal
166 wave-vector ($j = \sqrt{-1}$). Indices 0 and 1 refer to the MgO
167 substrate and the FeRh thin film, respectively. $D_i = \kappa_i / (\rho_i C_i)$
168 is the heat diffusivity with ρ_i and C_i the mass density and spe-
169 cific heat, respectively. $Q_f = \frac{4}{\pi} P_{\text{inc}} (1 - R)$ is the f -component
170 of the incoming modulated heat power with R the reflection
171 coefficient at the pump wavelength. J_0 is the Bessel func-
172 tion of the first kind, and α is the absorption coefficient of the
173 pump beam in FeRh³⁸. The last two terms of Eq. (1) describe
174 the finite diameter of the laser spots: $A(u)$ is the Hankel trans-
175 form of the Airy pattern of the diffraction-limited probe spot,
176 and d_{pump} is the diameter at which the intensity of the Gaus-
177 sian pump spot has fallen by e^2 . Numerical values for known
178 parameters are given in Appendix A .

179 The temperature increase includes *all* contributions to the
180 thermal conductivity: phononic, electronic, magnonic, as well
181 as the effect of any thermal interface resistance (TIR) R_{th} be-
182 tween the substrate and the thin film due to a localized scat-
183 tering of phonons. Taking into account the presence of a TIR
184 amounts to using in Eq. (3) a modified thermal conductivity
185 for the substrate $\kappa_0^* = \kappa_0 / (1 + R_{th} \kappa_0 \Sigma_0)$ ³⁹. We now discuss
186 possible values to give to R_{th} . The TIR is notoriously compli-
187 cated to measure reliably⁴⁰, particularly in the case of a ther-
188 mally insulating film/conducting substrate configuration. It
189 may have *intrinsic* origins such as *e.g.* Umklapp processes⁴¹,
190 or, more likely at play in our case, *extrinsic* origins such as
191 interface roughness - expected to be minute in our epitaxial
192 films. Applying the diffuse mismatch model to the FeRh/MgO
193 interface yields $R_{th} \sim 4 \times 10^{-9} \text{ m}^2 \text{ K W}^{-1}$ for both magnetic
194 phases (see below for numerical details on this simulation).
195 This method is known to give a lower bound for thermal in-
196 terface resistance, but matches reassuringly well with exper-
197 imental values found in the literature for thin metallic films
198 (e.g. Al or Cu) deposited on an electrically insulating sub-
199 strate (e.g. SiO₂ or Al₂O₃)^{42,43}. It is also quite close to
200 the value ($2 \times 10^{-9} \text{ m}^2 \text{ K W}^{-1}$) estimated indirectly for anti-
201 ferromagnetic FeRh/MgO by Ahn *et al.*¹⁹.

202 Modulated thermoreflectance spatial scans measured on
203 FeRh/MgO are typical of a thermal insulator over conducting
204 substrate configuration. In this case, heat diffuses rapidly to
205 the substrate, and then back to the top layer. To illustrate this,

TABLE I. Thermal conductivity extracted from group fits of the
log of the amplitude of the modulated reflectivity, imposing $R_{th} =$
 $4 \times 10^{-9} \text{ m}^2 \text{ K W}^{-1}$ as explained in the text.

Temperature (°C)	Phase	Thermal conductivity (W m ⁻¹ K ⁻¹)
5	AF	26 ± 3
40	AF	30 ± 5
60	AF	30 ± 5
100	FM	10 ± 1
130	FM	9 ± 1

206 we plot in Fig. 3(a) the amplitude of the temperature rise given
207 by Eq. (2) at an example frequency of $f = 250$ kHz, with and
208 without an FeRh layer ($h = 0$ or 195 nm). Far from the center,
209 the behavior of the MgO substrate is recovered, with the slope
210 essentially governed by the frequency-dependent ther-
211 mal diffusion length $\mu_0 = \sqrt{\frac{D_0}{\pi f}}$. The central temperature rise,
212 on the other hand, roughly scales as $\frac{h}{\kappa_1} + R_{th}$ above the signal
213 from the substrate (Fig. 3(a)). As a result, it is (i) quite chal-
214 lenging to measure reliably the thermal conductivity of very
215 thin/highly conductive films, and justifies the use of a fairly
216 thick (195 nm) film for this study, and (ii) almost impossible
217 to determine independently R_{th} and κ_1 , as the two terms al-
218 ways appear together⁴⁴. To analyze our data, we thus chose
219 to impose a value for R_{th} (see discussion above). Finally, we
220 verified that the $r = 0$ value of the amplitude scales linearly
221 with the incident power.

222 In order to evaluate the thermal conductivity of FeRh at a
223 given temperature, we record spatial scans of the amplitude
224 and phase of the modulated thermoreflectance signal at three
225 modulation frequencies (typically 100, 250 and 500 kHz), as
226 shown for instance for $T = 100^\circ \text{C}$ (373 K) in Fig. 3(b)-(c).
227 We then perform a global fit of the logarithm of the normal-
228 ized amplitude of the three curves, with the thermal conduc-
229 tivity of FeRh being the only free (shared) parameter and the
230 pump beam diameter and TIR imposed. Starting from an iden-
231 tical value of $R_{th} = 4 \times 10^{-9} \text{ m}^2 \text{ K W}^{-1}$ in both AF and FM
232 phases, we also explore the possibility of a much larger TIR
233 in the FM phase, and use the quality of the fit to validate the
234 chosen value. Note that no knowledge of $\frac{\partial R_{\text{probe}}}{\partial T}$ is necessary

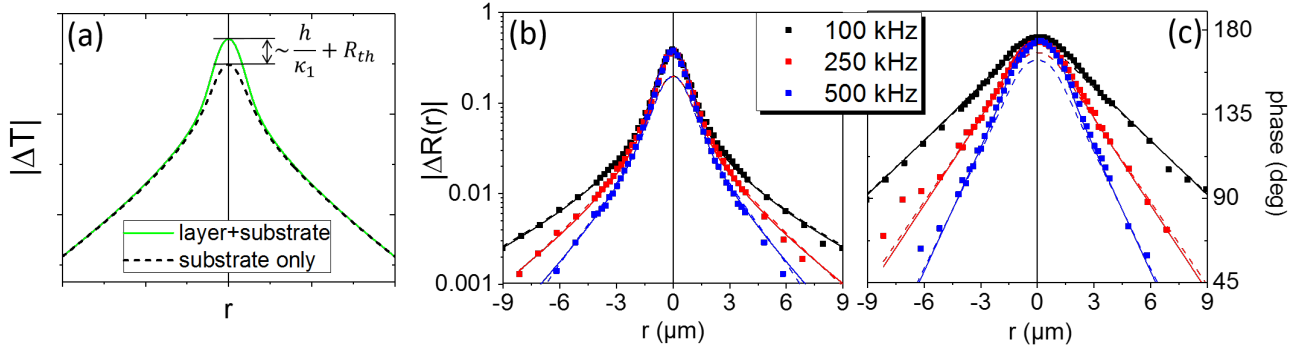


FIG. 3. (a) Illustration of the effect of the presence of a thin film over a better thermally conducting substrate : logarithm of the temperature rise induced by a 250 kHz modulated laser, with and without the 195-nm-thick FeRh film (Eq. 1) . The "nose" contains information on the layer's thermal conductivity and on the thermal interface resistance. (b-c) Experimental spatial scans of amplitude and phase taken at three modulation frequencies in the uniform FM phase ($T = 100^\circ\text{C}$, 373 K). The fit of $\log|\Delta R|$ by Eq. (1) yields $\kappa_1 = 10 \pm 1 \text{ W m}^{-1} \text{ K}^{-1}$. The response of the FeRh/MgO system (full line) and MgO substrate only (dashed line) are plotted using $d=2.1 \mu\text{m}$ and $R_{th} = 4 \times 10^{-9} \text{ m}^2\text{K W}^{-1}$.

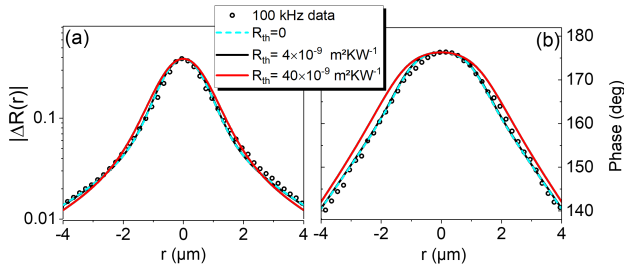


FIG. 4. (a) Amplitude and (b) phase at 100 kHz modulation frequency in uniform FM phase ($T = 100^\circ\text{C}$, 373 K). Fitting the data with different thermal interface resistances $R_{th}=0, 4$ or $40 \times 10^{-9} \text{ m}^2\text{K W}^{-1}$ gives $\kappa_1 = 9, 10$ or $20 \text{ W m}^{-1} \text{ K}^{-1}$ (resp. green, blue and red curves). The amplitude and phase are then calculated with Eq.(2). Note that the difference between $R_{th}=0$ and $40 \times 10^{-9} \text{ m}^2\text{K W}^{-1}$ curves is minute, and that the agreement with the phase data is poor for the larger TIR.

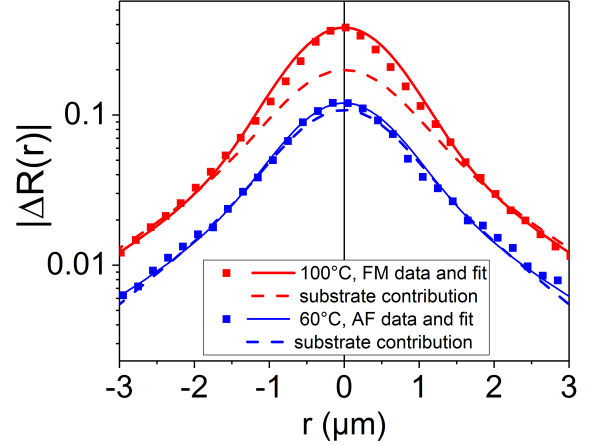


FIG. 5. Amplitude of the modulated reflectivity measured at 500 kHz in the uniform AF (resp. FM) phases at $T=60^\circ\text{C}$ in blue symbols (resp. $T=100^\circ\text{C}$ in red symbols). Full lines are fit to the data with $\kappa_1=30 \text{ W m}^{-1} \text{ K}^{-1}$ (AF) and $\kappa_1=10 \text{ W m}^{-1} \text{ K}^{-1}$ (FM), while the dashed line is the contribution of the MgO substrate.

235 to extract κ_1 from this analysis, since it is the relative spatial
 236 variations of the f -component of the reflectance that are of
 237 importance. Finally, complementary MatLab simulations including a third layer in the model confirm that we can neglect
 238 the presence of the thin Pt cap in the analysis of the data.
 239

240 Table I shows the final temperature dependence of the thermal
 241 conductivity of the 195-nm-thick FeRh film, established
 242 using the group fit procedure described above. The overall
 243 trend is distinctly that of a smaller value of $\kappa_1(T)$ in the FM
 244 phase compared to the AF phase. More specifically, κ_1 is 25-
 245 30 $\text{W m}^{-1} \text{ K}^{-1}$ in the AF phase, and around $10 \text{ W m}^{-1} \text{ K}^{-1}$
 246 in the FM phase, *i.e.*, almost three times less. This shows up
 247 clearly when plotting together spatial scans measured at the
 248 highest AF temperature (60°C) and lowest FM temperature
 249 (100°C), in Fig. 5. The "nose" of the AF spatial scan is strikingly
 250 smaller than in the FM scan. This could either be due to a
 251 smaller thermal conductivity in the FM phase, or to a much
 252 larger interface resistance. To test the latter hypothesis, we fit
 253 the FM data imposing a very large value of $R_{th} = 4 \times 10^{-8}$

254 $\text{m}^2\text{K W}^{-1}$ (red curve in Fig. 4). This does yield a higher
 255 $\kappa_{1,FM}$ of around $20 \text{ W m}^{-1} \text{ K}^{-1}$, but at the cost of a poorer fit
 256 quality, particularly visible on the phase (Fig. 4(b)). Note that
 257 this method sets an *upper* boundary to a likely value for R_{th} ,
 258 while DFT simulations give a lower limit.

259 We can now estimate the heat diffusivity $D_1 = \frac{\kappa_1}{\rho_1 c_1}$ of FeRh
 260 in both phases. Since the volume density of FeRh ρ_1 varies
 261 appreciably with the magnetic phase (AF or FM, see Ap-
 262 pendix A for numerical values), D_1 is divided by over two
 263 upon crossing the transition: from $\sim 8 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ in the
 264 AF phase at 5°C , to $\sim 3 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$ in the FM phase at
 265 130°C (403 K). Note that the former value is in the range of
 266 the diffusivity of $2 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ found by Bergman *et al.*⁸
 267 by fitting the laser-induced transient reflectivity, late after the

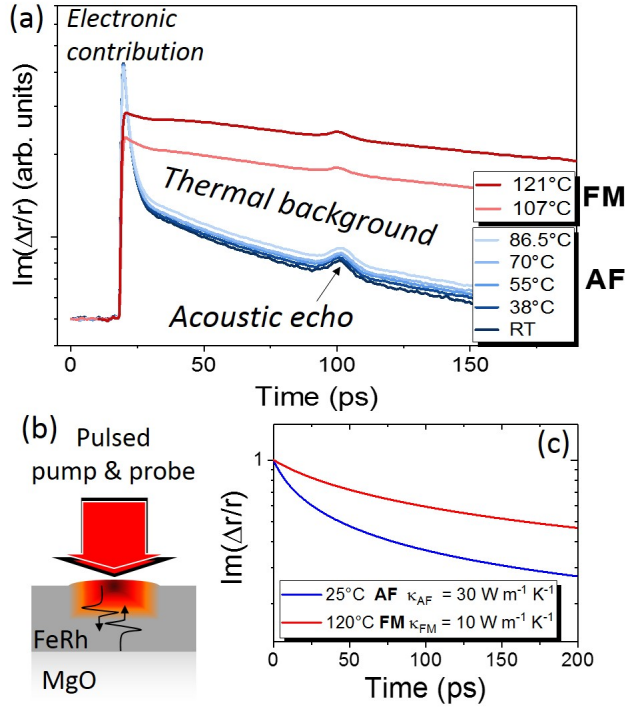


FIG. 6. (a) Interferometric signal at different temperatures on either side of the transition obtained using a time-resolved pump-probe setup (b)⁴⁵, with large diameter spots. Incoherent phonon generation occurs together with the launching of a thermo-elastically generated *coherent* strain wave that travels rapidly to the FeRh/MgO interface and back. The more rapid decay of the thermal background in the AF phase indicates a higher thermal conductivity as evidenced from (c) the calculated interferometric signal, given by Eq. 4 and normalized to its $t = 0$ value.

268 arrival of the pulse, when the layer is back into its AF phase.
 269 This makes FeRh roughly as heat-diffusive as MgO in its AF
 270 phase, but almost four times less in the FM phase. Typical
 271 thermal diffusion lengths at 500 kHz are then of a few microns,
 272 much more than the film thickness. A rough estimate
 273 in a 1D approximation moreover gives the typical time it takes

274 for the heat to escape laterally a $d \approx 2 \mu\text{m}$ diameter heat spot
 275 as $\tau \approx \frac{d^2}{D_1}$, i.e., $\tau_{AF} \approx 500 \text{ ns}$ while $\tau_{FM} \approx 1.2 \mu\text{s}$.

276 CONFIRMATION OF $\kappa_{AF} > \kappa_{FM}$ USING PULSED 277 GENERATION OF INCOHERENT PHONONS

278 The analysis of the thermoreflectance data points to a
 279 smaller thermal conductivity in the FM phase compared to the
 280 AF phase, even considering a larger thermal interface resistance
 281 at high temperature. This comes as a surprising result
 282 given that both the Wiedeman-Franz law and a previous exper-
 283 imental estimate¹⁹ seemed to indicate instead a larger thermal
 284 conductivity in the FM phase. To verify this, we performed pi-
 285 cosecond acoustics pump-probe experiments (see Fig. 6, and
 286 Appendix B for experimental details). A 80 MHz femtosec-
 287 ond laser beam is split in a train of pump pulses that gener-
 288 ate coherent strain waves by the thermoelastic effect, and de-
 289 layed probe pulses that measure the dephasing of the reflected
 290 electric field of light by interferometry⁴⁵. After an electronic
 291 signal decaying within 10-20 ps, one observes a slower decay
 292 related to the cooling of the layer as the heat is evacuated from
 293 the 12-13 nm thick top layer in which the light has been ab-
 294 sorbed (Fig. 6(a)). It is clearly slower in the FM compared to
 295 the AF phase, as it is also observed in the time-resolved strain
 296 measurements by Ahn *et al.* in Ref. 19 (see in particular
 297 their Supplemental Information⁴⁶). This indicates once more
 298 $\kappa_{1,AF} > \kappa_{1,FM}$. We can this time safely discard the influence of
 299 the interface resistance on the thermal background during the
 300 $t_{ps} = 300 \text{ ps}$ window of observation: whereas the longitudinal
 301 acoustic wave traveling at 4741 m s^{-1} easily sees the FeRh in-
 302 terface twice, the thermal wave will only have reached around
 303 $l_{th} = \sqrt{Dt_{ps}} \approx 30\text{-}50 \text{ nm}$ deep into the FeRh layer. The inter-
 304 ferometric signal is calculated as $\text{Im}(\Delta r(t)/r)$ where r is the am-
 305 plitude reflection coefficient of the light electric field (Eq. 4).
 306 The first term contains the surface displacement $u(t, z=0)$. k_0
 307 equal to $2\pi/\lambda$ is the light wavevector. The next term describes
 308 the reabsorption and dephasing of the reflected light taking
 309 into account the change of the refractive index $n = n' + jn''$
 310 induced by the propagating strain pulse S_{prop} and the temper-
 311 ature variation ΔT arising from heat diffusion.

$$\frac{\Delta r(t)}{r} = 2jk_0 u(t, z=0) + jk_0 \frac{4n}{(1-n^2)} \int_0^\infty \left(\frac{dn}{dS} S_{prop}(t, z) + \frac{dn}{dT} \Delta T(t, z) \right) \exp(j2k_0 n z) dz \quad (4)$$

312 The detailed expressions of $u(t, z=0)$, $S_{prop}(t, z)$ and $\Delta T(t, z)$
 313 and the parameters used in the modelling are given in Ap-
 314 pendix B. The surface displacement and propagating strain are
 315 found to give smaller contributions to the interferometric sig-
 316 nal as compared to heat diffusion. This signal is calculated
 317 using the thermal diffusivity given by κ values in Table I and
 318 shown in Fig. 6(c). It clearly evidences a faster decay in the
 319 AF phase with a larger diffusivity, in good agreement with the
 320 experimental results of Fig. 6(a).

DISCUSSION

322 Having confirmed the trend $\kappa_{1,AF} > \kappa_{1,FM}$ seen on the
 323 modulated thermoreflectance data, we compare the val-
 324 ues of the thermal conductivity in both phases (Table I),
 325 with that of other materials at room temperature. The
 326 absolute values are overall smaller than for⁴⁷ good met-
 327 als (a few hundreds $\text{W m}^{-1} \text{ K}^{-1}$), semiconductors ($\approx 10\text{-}$
 328 $100 \text{ W m}^{-1} \text{ K}^{-1}$), or even an electrical insulator such as MgO

329 ($\approx 50 \text{ W m}^{-1} \text{ K}^{-1}$). As often occurs upon alloying⁴⁸, the thermal conductivity of FeRh is also smaller than those of its
 330 constituting single elements^{47,49}: $\kappa_{Fe} \approx 80 \text{ W m}^{-1} \text{ K}^{-1}$ and
 331 $\kappa_{Rh} \approx 135 \text{ W m}^{-1} \text{ K}^{-1}$.

332 We can moreover estimate the stationary temperature increase due to the modulated green laser only, ΔT_{stat} , using Eq.
 333 (2) in which we set the frequency $f = 0$, $r=0$, and the incoming heat flow as $Q_0 = P_{inc}(1 - R)$. For an incident average
 334 laser power of $P_{inc} \approx 5.3 \text{ mW}$, the stationary temperature rise in the very center of a 2- μm -diameter pump spot will thus be
 335 of $\Delta T_{stat} \approx 13 \text{ }^\circ\text{C}$ for a base temperature of 40 or 60 $^\circ\text{C}$ (313 or 333 K) in the AF phase ($\kappa_1 \approx 30 \text{ W m}^{-1} \text{ K}^{-1}$). Note that this
 336 estimate is strongly dependent on the probe spot diameter.

342 CALCULATIONS OF κ_e , κ_{ph} , κ_m AND R_{th}

343 To dissect the experimentally observed and counter-intuitive thermal conductivity of FeRh, we perform simulations
 344 and combine with literature data to understand the individual phonon, electron and magnon contributions for the FM
 345 and AF phases.

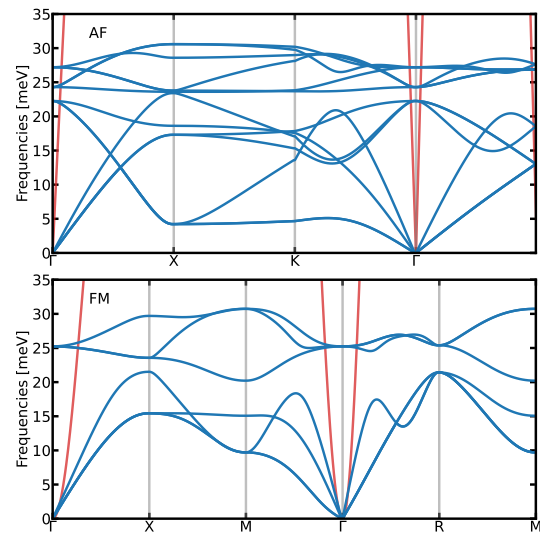
348 Phonon thermal conductivity: anharmonic phonon calculations

349 From an *ab initio* setting, the lattice contribution to the thermal conductivity can be computed by solving the phonon
 350 Boltzmann equation⁵⁰⁻⁵². In order to obtain quantitative results, it is essential to include effects beyond the harmonic
 351 approximation, such as thermal expansion and intrinsic anharmonicity, in the description of the system. This seems to
 352 be particularly true for the AF phase of FeRh, where previous theoretical studies predicted the appearance of an imaginary
 353 mode^{22,53-55}, which prevents the use of the harmonic approximation as a starting point for the Boltzmann equation. To
 354 go beyond the harmonic approximation, we employ the temperature dependent effective potential (TDEP) method^{52,56,57}
 355 to include anharmonicity and renormalize phonon-phonon interactions. To reduce the important simulation cost while
 356 keeping the accuracy of DFT, we constructed two machine-learning interatomic potential (MLIP⁵⁸⁻⁶⁰) models of FeRh
 357 for the FM and AF phases. The details of the simulations are described in Appendix C.

358 The resulting renormalized phonon band structures at room temperature are represented in Fig. 7. In the FM phase, all
 359 Fe atoms are equivalent by symmetry, so that the magnetic unit cell coincides with the B2 structure which can be used to
 360 compute phonon-related properties. This is not the case for the AF phase, where the spin up or down occupation of Fe
 361 atoms makes them inequivalent. Such a breaking of symmetry can have an important impact on the phonon properties²⁶.
 362 To account for this effect, the phonons in the AF phase were computed using the magnetic unit cell.

363 For both phases, we find that the spectra are fully real, demonstrating their dynamical stability. Some previous
 364 works^{22,24} had suggested the possibility of an additional low

380 T phase for the AF phase, which our results do not support (at
 381 least down to 50 K).



382 FIG. 7. Room temperature phonon band structure for (up) the AF and (down) the FM phase of FeRh. For comparison, the magnon
 383 band structures by Gu⁶¹ are also shown in red (assuming no magnetic anisotropy). The phonons in the FM phase are computed using the
 384 B2 structure as the unit cell, while the magnetic unit cell was used for the AF phase.

385 With these phonons and the third order phonon scattering as input, we obtained the lattice thermal conductivity of the
 386 two phases using the iterative solution of the Boltzmann equation. Our results give $\kappa_{ph,AF} = 11.84 \text{ W m}^{-1} \text{ K}^{-1} > \kappa_{ph,FM} =$
 387 $10.36 \text{ W m}^{-1} \text{ K}^{-1}$ at room temperature. Our calculation differs from previous work in the literature²⁶, where the opposite
 388 trend was found. The thermal conductivities computed by Cazorla and Rurali²⁶ were based on the harmonic approximation
 389 at the ground-state volume and using a different exchange and correlation functional. We show in Appendix C that
 390 beyond the differences in the DFT calculations that are minor, the main difference in our results comes from the explicit
 391 variation of the interatomic force constants and volume with temperature. The difference in lattice κ_{ph} favors the AF phase,
 392 but only slightly. However, the experimentally observed asymmetry between FM and AF thermal conductivities also contains
 393 electronic and magnonic contributions to κ , which we proceed
 394 to estimate.

402 Electron thermal conductivity

403 The electronic thermal conductivity κ_e can be estimated from the charge conductivity σ , through the Wiedemann-Franz
 404 relationship. The latter is known not to hold universally, but breakdowns mainly appear for strongly correlated
 405 metals, strange band structures, or sub-micron structures⁶². In the present case if the Lorenz factor takes its ideal value of
 406 $L_0 = 2.44 \times 10^{-8} \text{ W}\cdot\Omega\cdot\text{K}^{-2}$, we can extract the electron thermal conductivity as $\kappa_e = L_0 T \sigma$.

We measure the temperature-dependent resistance of a 195-nm-thick FeRh film which was grown together with the sample utilized for the thermoreflectance measurements above. The resistance measurement was carried out using the 4-point probe method by contacting the film with four pins arranged in a square 4 mm on each side. The temperature was controlled using a custom-made Peltier holder and no magnetic field was applied during the measurement. In this way we find at 360 K $1/\sigma_{AF} = 1 \mu\Omega \text{ m}$ and $1/\sigma_{FM} = 0.6 \mu\Omega \text{ m}$, very close to values from the literature⁶³. This implies $\kappa_{e,AF} \approx 9 \text{ W m}^{-1} \text{ K}^{-1}$ which is sensibly lower than $\kappa_{e,FM} \approx 15 \text{ W m}^{-1} \text{ K}^{-1}$. Theoretical calculations of κ_e in FeRh based on its electronic band structure also show a slightly larger value for the FM phase: Jimenez *et al.*²⁵ find $\kappa_{e,AF} \approx 21 \text{ W m}^{-1} \text{ K}^{-1}$ and $\kappa_{e,FM} \approx 27 \text{ W m}^{-1} \text{ K}^{-1}$, using a somewhat arbitrary temperature and energy independent relaxation time of 10^{-14} s . In their calculations the Lorenz factor is not presumed constant (and depends on the electronic band structure) but the κ_e is known only up to a rescaling by the relaxation time, which will be different for FM and AF phases, and should be energy/state dependent. Overall, we expect $\kappa_{e,FM}$ to be a bit larger than $\kappa_{e,AF}$, and of the order of $10\text{--}15 \text{ W m}^{-1} \text{ K}^{-1}$.

Magnon thermal conductivity: model dispersion and lifetime

Finally, we wish to estimate the magnon contribution to the thermal conductivity. Wu *et al.*²⁹ calculate simultaneous lattice and spin dynamics for BCC Fe, and find that the magnon contribution to κ ($\kappa_m = 15 \text{ W m}^{-1} \text{ K}^{-1}$) is larger than the lattice one ($\kappa_{ph} = 8 \text{ W m}^{-1} \text{ K}^{-1}$), though they are skeptical of their κ_m . We have re-implemented the magnon band model from Gu and Antropov⁶¹ as a python script, and added the calculation of the group velocities $v_{q\lambda}$ and a lifetime given by $1/\tau_{q\lambda} = \alpha_G \omega_{q\lambda}$ which depends only on the frequency of mode λ at wave vector q . Here α_G is the Gilbert damping parameter, which should vary with temperature and magnetic ordering (and in principle also with the magnon mode λ, q). Using these ingredients, one can calculate the thermal conductivity with a formula analogous to that for phonons (Eq. (11) in Appendix C):

$$\tilde{\kappa}_m^{ab}(\omega) = \frac{1}{V} \sum_{q\lambda} v_{q\lambda}^a v_{q\lambda}^b C_v(\omega_{q\lambda}) \tau_{q\lambda} \delta(\omega - \omega_{q\lambda}) \quad (5)$$

$$\kappa_m^{ab} = \int \tilde{\kappa}_m^{ab}(\omega) d\omega \quad (6)$$

where a, b are Cartesian components, C_v is the mode specific heat ($[1/2x/(\sinh x/2)]^2$ with $x = \hbar\omega/k_B T$), and V is the unit cell volume. We call $\tilde{\kappa}$ the spectral thermal conductivity. The q integrations are carried out numerically on 100^3 point grids for the primitive cubic Brillouin Zone. In this framework, the band structure strongly favors $\kappa_{m,FM}$, through the much larger DOS at low frequency (quadratic vs linear dispersion), which boosts the C_v at low ω , where τ is also large. This effect is stronger than that of the magnon group velocities, which favor the AF phase (constant acoustic velocity), over the FM phase (zero velocity at Γ which grows linearly). The product

$C_v \tau v^2$ yields $\tilde{\kappa}_{m,FM} \propto \omega^{1/2}$ for the FM phase, while $\tilde{\kappa}_{m,AF} \propto \omega$. The Bose distribution weights most of C_v and κ_m at low frequencies, generically favoring $\kappa_{m,FM}$ over $\kappa_{m,AF}$.

An important ingredient of the calculation is the parameter α_G , in both phases. The only measurements of FeRh magnon damping have been performed in the uniform FM phase, for which quite a large spread has been reported. Intrinsic damping values of around 0.001–0.004 have been measured⁶⁴ or computed using a multiple scattering method including position and spin fluctuations⁶³. Larger values between 0.03 and 0.1 have also been seen, and attributed to the spin-sink effect of either an adjacent Pt layer⁶⁵, or residual AF domains⁶⁶.

For the AF phase, a first principles calculation was made by Mahfouzi and Kioussis⁶⁷, and using their Eq. 5 we obtain values ranging from 0.03 to 0.3 between what they call low and high temperature. Simensen *et al.*⁶⁸ arrive at 0.3 with a magnon scattering theory.

Using a single conservative value $\alpha_G = 0.25$ for both phases, we obtain values of $\kappa_{m,AF} \sim 15 \text{ W m}^{-1} \text{ K}^{-1}$ and $\kappa_{m,FM} \sim 90 \text{ W m}^{-1} \text{ K}^{-1}$. The former is reasonable, but the latter is too large compared to our measurements. Examining the different ingredients for our model, the magnon dispersion compares favorably with experiments⁶⁹ and the calculations of Sandratskii and Buczek⁷⁰, except for a small but crucial underestimation of the FM phase stiffness at low frequency and q , noted by Castets⁶⁹. If the acoustic mode has a slight linear component (e.g. by mixing with phonon excitations) or much higher stiffness, the DOS, C_v and $\kappa_{m,FM}$ will be strongly reduced. The main source of uncertainty is the simplistic lifetime model as $1/\omega\alpha_G$: the large DOS and lifetimes at low frequency give too much weight to the FM phase κ_m .

Estimate of the thermal resistance at the FeRh/MgO interface

The thermal interface resistance was estimated theoretically based on the bulk phonon dispersions of FeRh (FM and AFM) combined with those of MgO. The diffuse mismatch model (DMM) was used^{71,72}, which has two main hypotheses: (i) the phonons are transmitted based on their frequency matching and their group velocity component normal to the interface; (ii) detailed balance and a steady state is achieved, which allows to calculate the transmission probability. The implementation was carried out in the Abinit⁷³ package following the full phonon description in Ref.⁷⁴ and was benchmarked against their values for interfaces between Si, Cu and Al. For (100) oriented FeRh on (100) MgO⁷⁵, the AFM phase has a room temperature interface resistance of $4.3 \times 10^{-9} \text{ m}^2 \text{ K W}^{-1}$, and the FM phase of $4.2 \times 10^{-9} \text{ m}^2 \text{ K W}^{-1}$. Other crystalline orientations tend to decrease the resistance, down to 2.3 (resp. 0.24) $\times 10^{-9} \text{ m}^2 \text{ K W}^{-1}$ for FM (resp. AF) (111) FeRh on (111) MgO. The main factor boosting the interface resistance is the small overlap in frequency between light MgO and heavier FeRh. To fit the thermoreflectance spatial scans, we used the value $R_{th} = 4 \times 10^{-9} \text{ m}^2 \text{ K W}^{-1}$, but also considered the possibility of much larger values (see Fig. 4).

TABLE II. Numerical estimates of the different contributions to the thermal conductivity. The last column refers to the total value, to be compared to the experimentally measured one (Table I).

Phase	κ_{ph}	κ_e	κ_m	κ (W m ⁻¹ K ⁻¹)
AF	12	9	15	36
FM	10	15	90	115

DISCUSSION AND COMPARISON TO EXPERIMENTS

We find three comparable contributions for the thermal conductivity, between the lattice, electronic, and magnon terms (Table II). Each term has been estimated using available simulations or experimental input, and they are all on the order of 10 W m⁻¹ K⁻¹, with $\kappa_{m,FM}$ the only outlier. The sum renders quite nicely the measurements of $\kappa_{AF} \sim 30$ W m⁻¹ K⁻¹, but the lower measured κ_{FM} requires additional attention: the observed difference in the lattice κ_{ph} provides 1–2 W m⁻¹ K⁻¹, but the other two terms favor FM over AF.

Several factors may contribute to lower κ_{FM} :

- the multi-domain state of the layer in its FM phase could give an extra contribution from ferromagnetic domain-wall resistance⁷⁶, but a large number of ferromagnetic domains is quite unlikely under the two-micron spot, due to the competition between exchange and magnetostatic energy cost.
- disorder will limit transport in both phases, but magnetic disorder (e.g., domains, impurities) will affect the FM phase transport more strongly as the dipolar interaction is longer ranged. The strong κ_m advantage of FM over AF for long wavelengths will be reduced by defects and grain boundaries.
- the low- q magnon phonon coupling is bound to play a critical role (see Fig. 7). Due to their quadratic dispersion and very similar frequencies, the FM magnons will interfere with acoustic phonons, limiting both lattice κ_{ph} and magnon κ_m . The impact of this coupling on the latter could have a significant impact on the total thermal conductivity on the FM phase. Compounding this effect, the AF magnon group velocity is much higher and there will be no coupling of magnons to acoustic phonons.
- Our model for magnon lifetimes is quite crude, and may break down. Beyond the temperature and magnetic state, the “true” $\alpha_{G,q\lambda} = 1/\omega_{q\lambda} \tau_{q\lambda}$ will also depend on the specific magnon mode. Any upper bound on the lifetimes at low q and low frequency will again reduce κ_{FM} more strongly.

CONCLUSIONS

Space and frequency-dependent thermorefectance measurements were performed in order to determine the thermal

conductivity of a 195-nm-thick FeRh film in a wide temperature range of 5 to 130°C, across the AF to FM transition. Unexpectedly (based on rough estimates made with the Wiedemann-Franz law), the thermal conductivity is found overall three times *larger* in the AF phase compared to the FM phase, a trend confirmed by the temporal decay of laser-pulsed generated incoherent phonons. These values were then used to estimate the expected transient and stationary temperature rises induced by a laser. To explain why the total FM thermal conductivity is lower than the AF one, we estimated the phononic contribution to κ from anharmonic first-principles dynamics, the electronic contribution from the experimental resistivity using Wiedemann-Franz, and the magnonic contribution using a linear spin wave model from the literature, with a Gilbert-type relaxation time. In the AF phase, the three components were found to be of the same order of magnitude, with a total of around 36 W m⁻¹ K⁻¹, very close to the observed ≈ 30 W m⁻¹ K⁻¹. In the FM phase, however, the calculated thermal conductivity rockets to ≈ 115 W m⁻¹ K⁻¹, which is unrealistically large with regards to the observed ≈ 10 W m⁻¹ K⁻¹. More accurate modelling of electronic and magnonic contributions will be required: in particular the large uncertainties in the magnetic damping, the magnon-phonon coupling, and the effects of disorder lead to a strong overestimate of all three components of κ .

These results underscore the complexity of FeRh and the importance of the three-way couplings between electrons, phonons, and magnons, which are on equal footing in the total thermal conductivity.

ACKNOWLEDGMENTS

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APPENDIX A: PARAMETERS

The values for the thermal conductivity $\kappa_0(T)$, diffusivity $D_0(T)$ and specific heat $C_0(T)$ of MgO were taken from

606 Ref. 77. The specific heat $C_1(T)$ of FeRh was found in 626
 607 Ref. 1, where they measured much thicker, polycrystalline 627
 608 films. We believe any error on this parameter should impact
 609 only mildly the determination of κ_1 . Indeed this par-
 610 ameter intervenes through the calculation of the diffusivity
 611 of FeRh, $D_1(T)$, which has a minor effect on the analysis of
 612 modulated thermoreflectance experiments in the "insulating-
 613 over-conductive layer" configuration³⁰. The volume den- 628
 614 sity of FeRh was taken phase-dependent and temperature- 629
 615 independent⁷⁸ from Ref. 22: $\rho_{1,AF} = 9744.51 \text{ kg m}^{-3}$ and 630
 616 $\rho_{1,FM} = 9590.11 \text{ kg m}^{-3}$. A standard knife-edge measure- 631
 617 ment in our experimental geometry gives $d_{\text{pump}} = 2. \pm 0.1 \mu\text{m}$. 632
 618 The reflectance of FeRh was taken phase-dependent and esti- 633
 619 mated as: $R_{AF} = 0.75$ and $R_{FM} = 0.72$ by comparing to the 634
 620 reflectance of Aluminium. Finally, the light absorption coef- 635
 621 ficient of FeRh was estimated in both phases by measuring 636
 622 the light reflected from/transmitted through a 30 nm sample 637
 623 grown in similar conditions. It corresponds to a penetration 638
 624 depth of around 10 nm (resp. 12-13 nm) at 532 nm (resp. 639
 625 773 nm), much less than the thickness of the film $h = 195 \text{ nm}$. 640

APPENDIX B: DETAILS ON THE PICOSECOND ACOUSTICS EXPERIMENTS AND MODELLING

628 The picosecond acoustics pump-probe set-up is described
 629 in Peronne *et al.*⁴⁵. More specifically to these measurements,
 630 the laser repetition rate was 80 MHz, with a modulation of
 631 1 MHz. Its wavelength was 773 nm, and the beam diameter
 632 was of the order of $\approx 15 \mu\text{m}$. The power of the pump beam
 633 was around $P=32 \text{ mW}$, that of the probe around 4 mW. The
 634 delay line was scanned mechanically at 40 nm/ps. The model
 635 is based on the 1D heat diffusion equation and the propagation
 636 equation for elastic waves. Given the short time scale (300 ps),
 637 the FeRh layer can be considered as a semi-infinite medium
 638 since the thermal wave has not reached the substrate. The
 639 surface displacement $u(t, z = 0)$, propagating strain $S_{\text{prop}}(t, z)$
 640 and temperature increase $\Delta T(t, z)$ are obtained as⁷⁹:

$$u(0, t) = -\frac{S_0}{\alpha} \left(1 - \frac{x \exp(-\omega_\alpha t)}{x+1} - \frac{x \exp(\omega_D t)}{x^2-1} \text{Erfc}(\sqrt{\omega_D t}) + \frac{\exp(x\omega_\alpha t)}{x^2-1} \text{Erfc}(\sqrt{x\omega_\alpha t}) \right) \quad (7)$$

$$S_{\text{prop}} = \begin{cases} z - vt > 0 & \frac{S_0}{1-x^2} (\exp(-\omega_D(z/v-t)) - x \exp(-\omega_\alpha(z/v-t))) \\ z - vt < 0 & \frac{S_0}{1-x^2} (\exp(\omega_D(t-z/v)) \text{Erfc}(\sqrt{\omega_D(t-z/v)}) - x \exp(x\omega_\alpha(t-z/v)) \text{Erfc}(\sqrt{x\omega_\alpha(t-z/v)}) \end{cases} \quad (8)$$

$$\Delta T(t, z) = \frac{F(1-R)\alpha}{2\rho_1 C_1} \exp(\omega_D t) \left(\exp(-\alpha z) \text{Erfc}\left(\frac{-z + 2D_1 \alpha t}{2\sqrt{D_1 t}}\right) + \exp(\alpha z) \text{Erfc}\left(\frac{z + 2D_1 \alpha t}{2\sqrt{D_1 t}}\right) \right), \quad (9)$$

641 where α , D_1 , v , ρ_1 , C_1 , R , are the absorption coef- 661
 642 ficient, diffusivity, longitudinal acoustic velocity, specific 662
 643 heat, and reflectivity coefficient of FeRh, respectively. 663
 644 We take $\kappa_{AF}(\kappa_{FM})=30(10) \text{ W m}^{-1} \text{ K}^{-1}$, hence $D_{AF}(D_{FM}) =$ 664
 645 $9.3(2.7) \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$, and $v_{AF}(v_{FM})=4741(4865) \text{ m s}^{-1}$. 665
 646 $F=0.2 \text{ mJ cm}^{-2}$ is the laser fluence. We note $\omega_\alpha = \alpha v$, 666
 647 $\omega_D = D_1 \alpha^2$, $x = \omega_\alpha / \omega_D$. The real and imaginary parts 667
 648 of the refractive index $n = n' + jn''$ were obtained at 668
 649 $\lambda = 773 \text{ nm}$ as a function of temperature by ellipsometry 669
 650 from 20°C to 120°C. For instance at 25°C for the AF 670
 651 phase and 120°C for the FM phase: $n'_{AF}(n'_{FM})=4.1(4.2)$, 671
 652 $n''_{AF}(n''_{FM})=5(4.4)$ and hence $\alpha_{AF}(\alpha_{FM})=8.1(7.2) \times 10^7 \text{ m}^{-1}$. 672
 653 The derivatives are $dn'_{AF}/dT(dn'_{FM}/dT) = 7.6(6.8) \times 10^{-3}$ 673
 654 and $dn''_{AF}/dT(dn''_{FM}/dT) = 4(2.8) \times 10^{-3}$. S_0 is defined as 674
 655 $3\beta B \alpha (1-R)F / (\rho_1^2 C_1 v^2)$ where β is the thermal expansion 675
 656 coefficient, and B the bulk modulus. Using data from^{19,80} we 676
 657 have $S_{0AF}(S_{0FM}) = 3.8(2) \times 10^{-4}$. The derivative dn/dS is 677
 658 not known but taking a large value of 30 consistent with data 678
 659 from the literature⁸¹, we get a smaller contribution of the prop- 679
 660 agating strain in the interferometric signal.

APPENDIX C: METHOD AND COMPUTATIONAL DETAILS FOR LATTICE THERMAL CONDUCTIVITY

Temperature dependent effective potential

664 The temperature dependent effective potential^{52,56,57}
 665 method is based on the construction of an effective anhar-
 666 monic Hamiltonian of the form

$$H = \sum_i \frac{p_i^2}{2M_i} + \frac{1}{2} \sum_{ij} \sum_{ab} \Theta_{ij}^{ab} u_i^a u_j^b + \frac{1}{3!} \sum_{ijk} \sum_{abc} \Psi_{ijk}^{abc} u_i^a u_j^b u_k^c \quad (10)$$

667 where u_i^a and p_i are respectively the displacement along the
 668 cartesian direction a and momentum of atom i with mass M_i ,
 669 and Θ_{ij}^{ab} and Ψ_{ijk}^{abc} are the second and third order interatomic
 670 force constants, which we fit to molecular dynamics data us-
 671 ing ordinary least-squares methods. The effectiveness of this
 672 Hamiltonian comes from the iterative nature of the fit, where
 673 each order is fit on the residual of the previous order. While
 674 this procedure ensures that the lowest orders intrinsically in-
 675 clude most of the anharmonicity, it also produces, as the name
 676 suggests, a temperature-dependent effective potential, so that
 677 individual MD simulations have to be run for each tempera-
 678 ture of interest.

679 Once the interatomic force constants are extracted from

680 MD simulations, the thermal conductivity can be computed
 681 using the Boltzmann equation. In this work, the solution of
 682 this equation was obtained with an iterative algorithm⁵⁰, after
 683 which the thermal conductivity tensor is written as

$$\kappa_{ph}^{ab} = \frac{1}{V} \sum_{\lambda} c_{\lambda} v_{\lambda}^a F_{\lambda}^b \quad (11)$$

684 with V the volume of the unit cell, c_{λ} the heat capacity asso-
 685 ciated with mode λ , v_{λ}^a the group velocity of mode λ along
 686 cartesian direction a , and F_{λ}^a is the non-equilibrium phonon
 687 distribution accounting for the phonon relaxation, for which
 688 the BTE must be solved self-consistently. The Boltzmann
 689 equation was solved on a $25 \times 25 \times 25$ \mathbf{q} -point grid and us-
 690 ing an adaptive gaussian scheme for the Dirac delta⁸².

691 For both the extraction of the interatomic force constants
 692 and the computation of the thermal conductivity, we used the
 693 implementation provided by the TDEP package⁸³.

694 Machine-learning interatomic potential and Molecular 695 dynamics

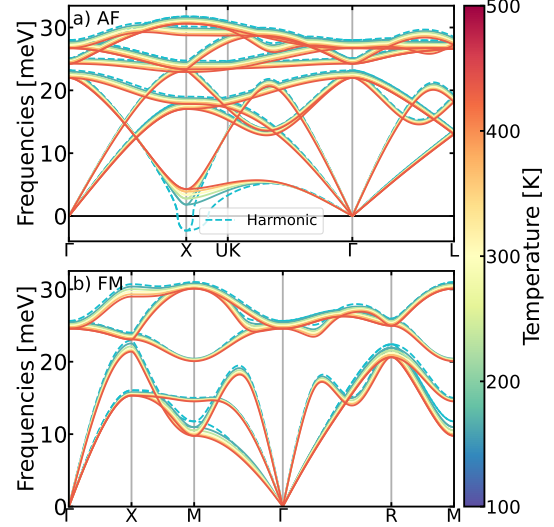
696 To decrease the important computational cost associated
 697 with the MD simulations, we used two machine-learning
 698 interatomic potential (MLIP) within the Moment Tensor
 699 Potential⁵⁹ formalism, one for the AF phase and the other
 700 one for the FM phase. The MLIP were fit on DFT calcula-
 701 tions performed with the Abinit suite⁷³ using the PBE param-
 702 eterization of the exchange and correlation functional⁸⁴ in the
 703 PAW formalism^{85,86}. These MLIP were successfully used to
 704 study the elastic properties of FeRh and we refer to our previ-
 705 ous work⁸⁰ for more details about the parameters and dataset
 706 used in their construction.

707 With the MLIP, we compute the effective anharmonic
 708 Hamiltonian from 100 to 500 K in steps of 100 K. For each
 709 temperature, we run two 100 ps MD simulations on $8 \times 8 \times 8$
 710 supercells, with a time step of 1 fs using the LAMMPS
 711 package⁸⁷. The first MD run is performed in the NPT en-
 712 semble, and is used to compute the average equilibrium vol-
 713 ume, while the second one employs this equilibrium volume
 714 in the NVT ensemble, in order to compute the renormalized
 715 interatomic force constants. These force constants are com-
 716 puted using 900 uncorrelated configurations, extracted from
 717 the NVT MD trajectory after 10 ps of equilibration.

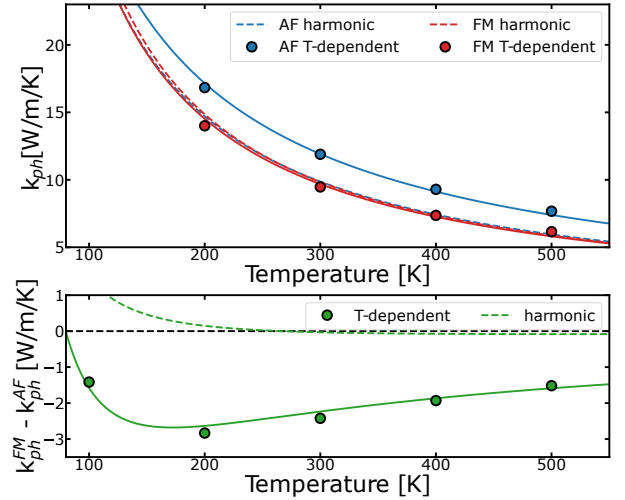
718 Temperature dependent phonons

719 The temperature dependent phonon dispersions of both
 720 phases are plotted on Fig. 8. For the FM phase, we ob-
 721 serve only a slight softening of the phonon frequencies with
 722 an increase in temperature, indicating a relatively small an-
 723 harmonicity in the system even at a temperature of 500 K.
 724 It should be noted that most of this temperature dependence
 725 can be attributed to the thermal expansion. While most of the
 726 modes in the AF phase show similar behavior, this is not the
 727 case for the lowest energy mode located at the high symmetry

728 X point. This mode, which we find to be imaginary in the har-
 729 monic approximation (as in previous literature, e.g.,^{22,53–55})
 730 has instead the opposite trend: a strong hardening with tem-
 731 perature. We note that even at 100 K, which is the lowest
 732 temperature studied here, this mode and the whole spectrum
 733 are fully real, demonstrating the stability of the AF phase.



734 FIG. 8. Temperature dependent phonons for a) the FM and b) the AF
 735 phase of FeRh. In b) the harmonic phonons computed with finite dif-
 ference to highlight the stabilization of the system by anharmonicity.
 The phonons in the FM phase are computed using the B2 structure as
 the unit cell, while the magnetic unit cell was used for the AF phase.



736 FIG. 9. a) Theoretical lattice thermal conductivity of FeRh computed
 737 for the AF phase, in blue, and the FM phase, in red. The harmonic
 738 results, represented with dashed lines, are obtained with IFC com-
 puted at $T = 25$ K. b) Difference of thermal conductivity between the
 FM and the AF phases.

736 Using these force constants as input, we computed the ther-
 737 mal conductivity by solving the iterative Boltzmann equation
 738 for each temperature. Our results, Fig. 9, show a slightly

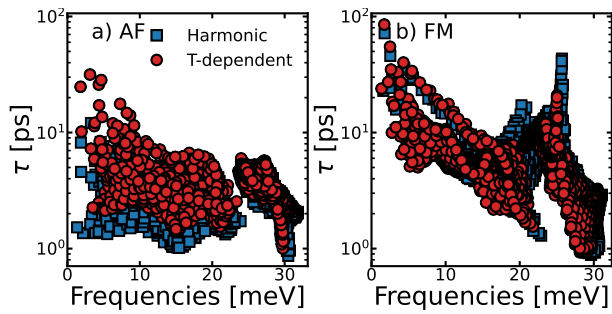


FIG. 10. Phonon lifetimes at 300 K in a) the AF phase and b) the FM phase. In both figures, the harmonic results are taken without accounting for the temperature dependence of the phonons.

larger κ_{ph} for the AF phase than for the FM phase above 150 K. As already stated in the main text, these finding differs from the results of Cazorla and Rurali²⁶, where the opposite trend was found based on the harmonic approximation at the

OK relaxed volume. To better understand the origin of this discrepancy, we computed the thermal conductivities with our MLIP using analogous approximations. Due to the imaginary mode in the AF phase, and in order to provide a meaningful comparison, we used IFC computed at a temperature of 25 K and at the reference relaxed volume for both phases, instead of the harmonic ones. As shown in Fig. 9, these approximations yield the same trend as in Ref. 26, with $\kappa_{ph,FM} > \kappa_{ph,AF}$ for all temperatures. To explain the opposite trend obtained with fully anharmonic IFC, we compare the $T = 300$ K phonon lifetimes obtained with the two approaches in Fig. 10.

The FM phase presents only a slight evolution of the lifetimes, confirming the low anharmonicity of this phase. On the contrary, the renormalization of the IFC brings an important *increase* of the phonon lifetimes in the AF phase. This counter-intuitive effect can be understood as the decrease of the scattering phase space due to the renormalization of the phonons⁸⁸. In the AF phase of FeRh, this effect is sufficiently strong for $\kappa_{ph,AF}$ to exceed $\kappa_{ph,FM}$.

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