



Laser-Assisted Antiferromagnetic-Ferromagnetic Transition in FeRh

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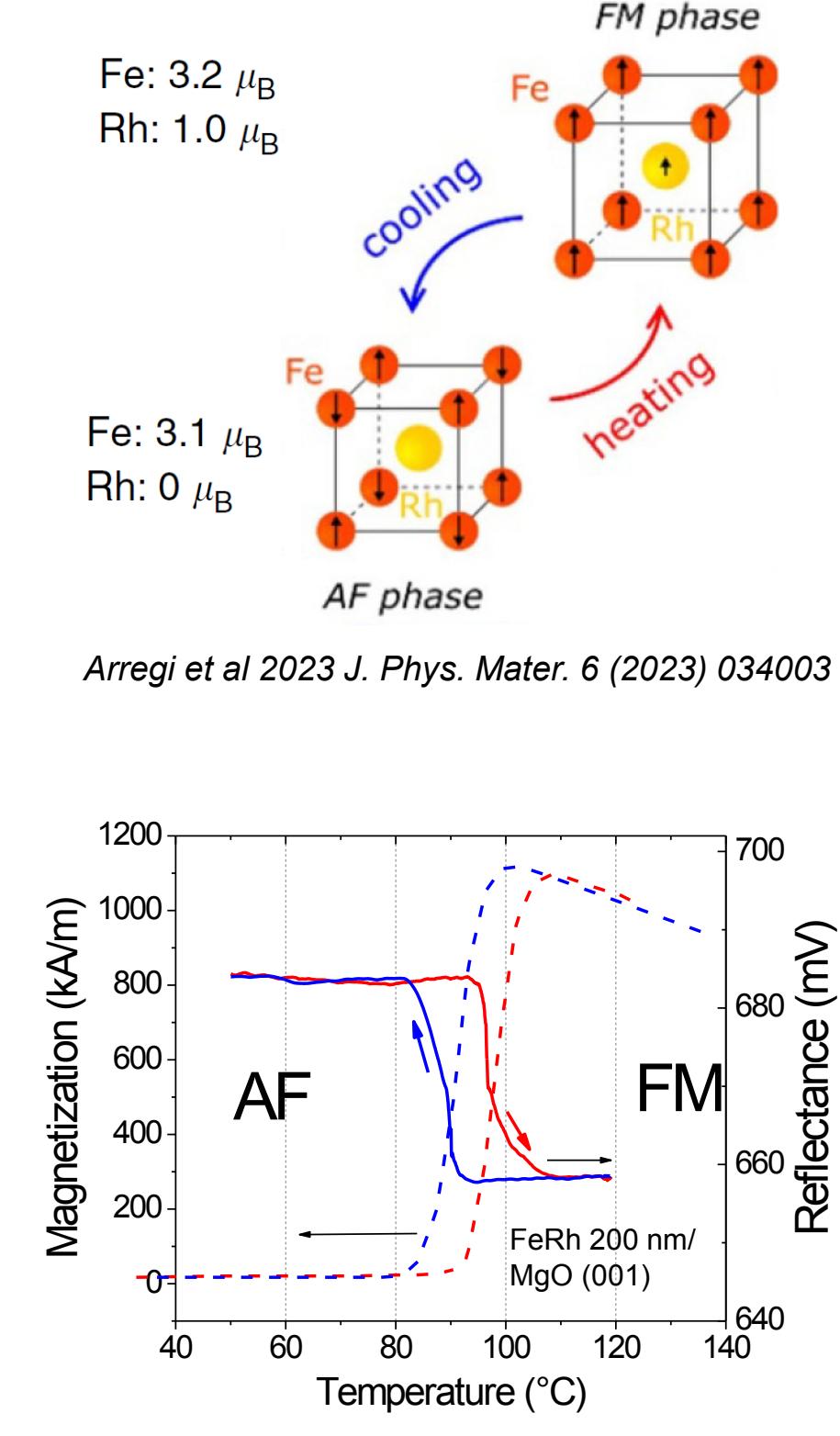
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Among magnetic materials near equi-atomic FeRh alloys, with their antiferromagnetic (AF) to ferromagnetic (FM) magneto-structural transition near room temperature, were studied long ago in bulk form [Kouvel, JAP 1962, Lewis, J. Phys. D 2016]. In the recent years they have aroused a renewed interest as thin films with particular focus on the tuning of their AF-FM transition [Arregi, J. Phys. D: 2018, Ceballos, APL 2017], the electrical detection of their magnetic state with application to information coding [Marti, Nat. Mater., 2014], and their giant magneto-caloric effect [Nikitin, Phys. Lett. 1990, Manekar, J. Phys. D 2008].

FeRh is also a good candidate to investigate laser-assisted transient or irreversible first order phase transition. Lattice expansion, nucleation of FM domains and magnetization dynamics have been studied using ultrafast pump-probe techniques. Here we show that low frequency (~ 100 kHz) modulated reflectance experiments provide an insight, not only on thermal effects (thermoreflectance, determination of the thermal conductivity) but also on the laser-assisted AF-FM domain movement and reversible/irreversible nucleation of the FM phase across the full temperature range of the AF-FM transition.

1 FeRh: AF to FM transition

- Fe₅₀Rh₅₀: AF-FM transition close to room temperature
- isostructural transition but increase of the lattice parameter by 0.7 %
- change of the electronic structure
 - optical properties: 4% decrease of reflectivity from AF to FM
 - electrical properties: 40% drop of resistivity from AF to FM
- transition temperature tunable by
 - strain
 - magnetic field
- giant magnetocaloric effect
- information coding on two perpendicular AF states
- laser-induced transient phase transition (Ju et al., PRL 2004, Bergman, PRB 2006, Radu PRB 2010, Mariager, PRL 2012, Pressacco, Str. Dyn. 2018, Nat. Commun. 2021, Li, Nat. Com. 2022, Mattern Arxiv 2023)

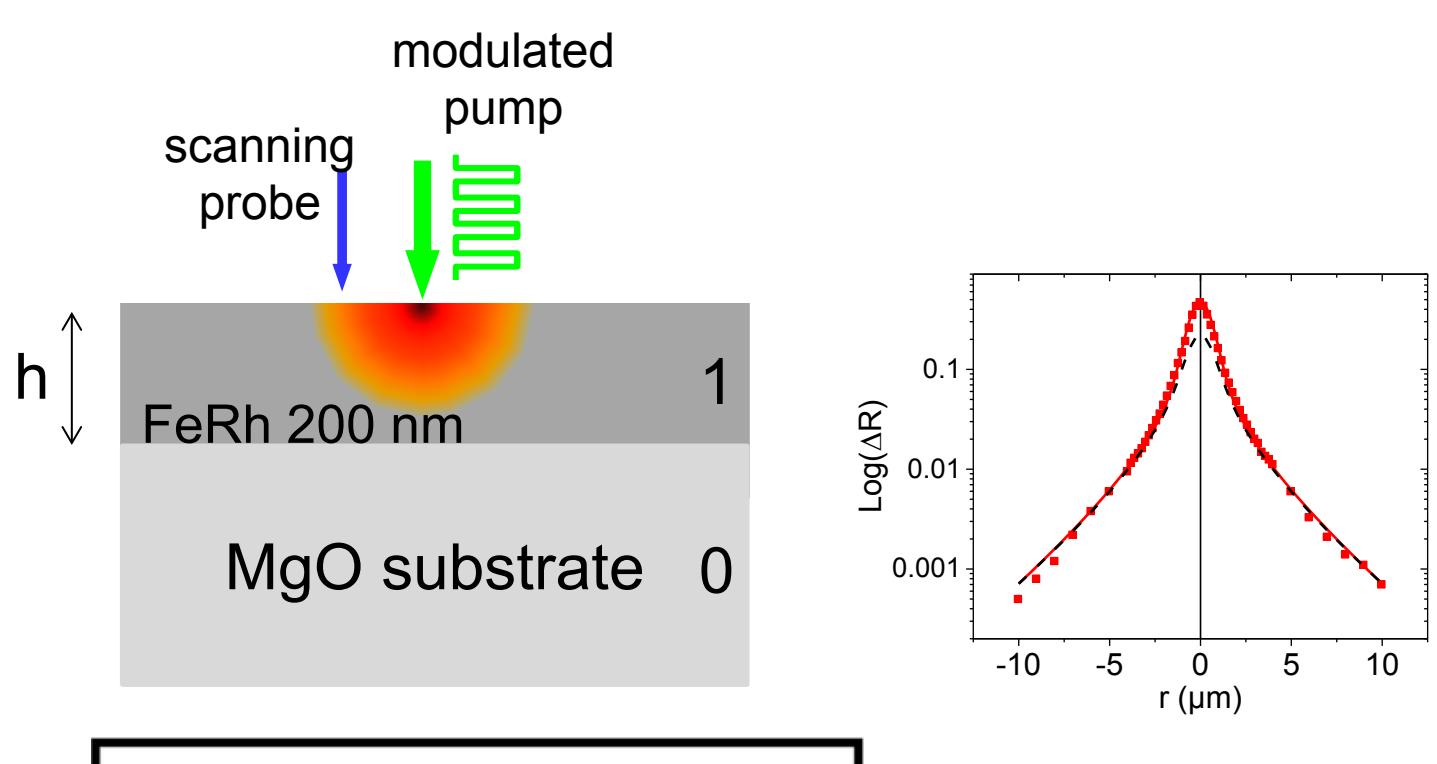


- time domain fs-ps-ns
- interplay between electron-phonon-magnon
- nucleation of FM phase+domain growth+ building of magnetization+magnetization dynamics
- DC + frequency domain
- only thermal effects
- local: μm spot
- determination of thermal conductivity and diffusivity
- nucleation and slow domain wall movement followed across the transition

epitaxial FeRh films onto single-crystal MgO (001) substrates post-growth annealing to obtain the CsCl-type (B2 ordering) crystal structure cap layer Pt 2 nm

Sample growth at CEITEC by DC magnetron sputtering (V. Uhlíř et al Nat. Commun. 7, 13113, (2016))

2 Modulated reflectance



$$R = R(T_0) + \Delta R_{thermo}$$

$$R = R(T_0) + \frac{\partial R}{\partial T} \Delta T$$

thermoreflectance

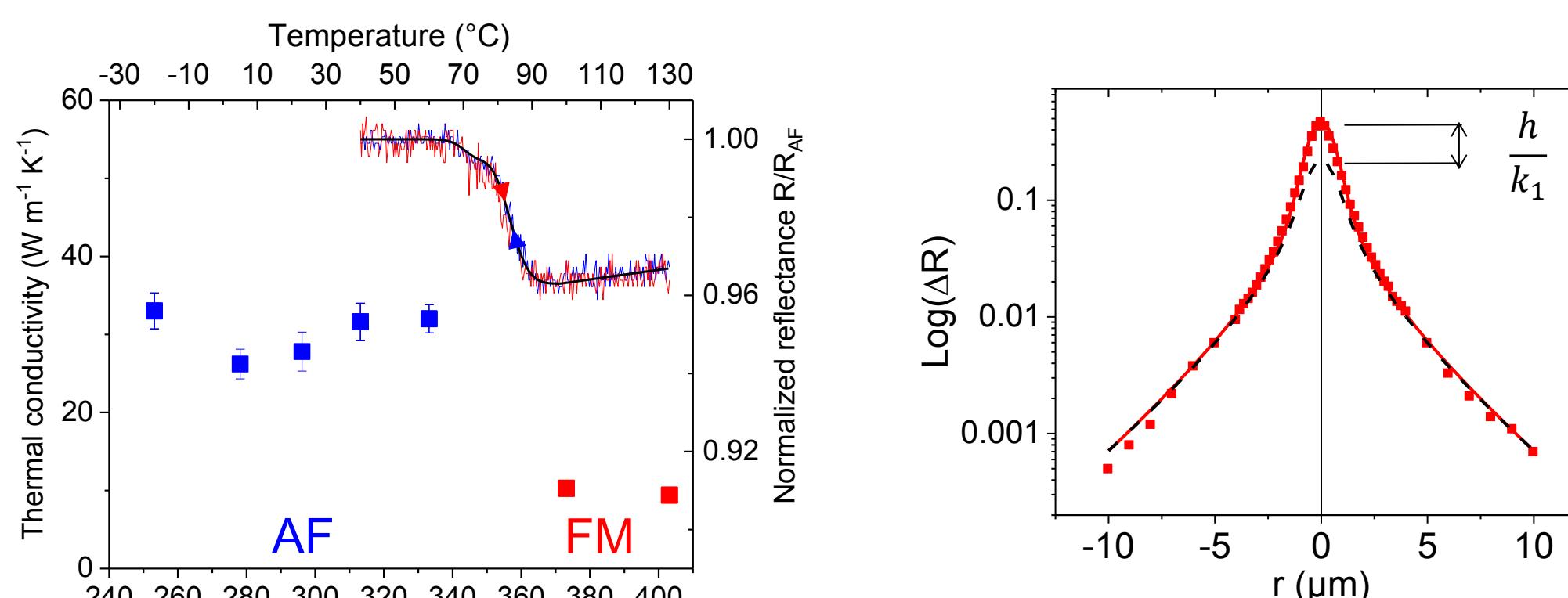
Heat diffusion in cylindrical geometry + boundary conditions

$$\Delta T_w(r) \approx e^{i\omega t} \frac{P(1-R)}{2\pi} \int_0^\infty \left(\frac{1}{k_0 \sigma_0(u)} + \frac{h}{k_1} \right) \exp\left(-\frac{d^2 u^2}{32}\right) J_0(u r) u du$$

$\sigma(u) = \sqrt{u^2 + \frac{i\omega}{D}}$ substrate layer Hankel transform of the Gaussian pump spot

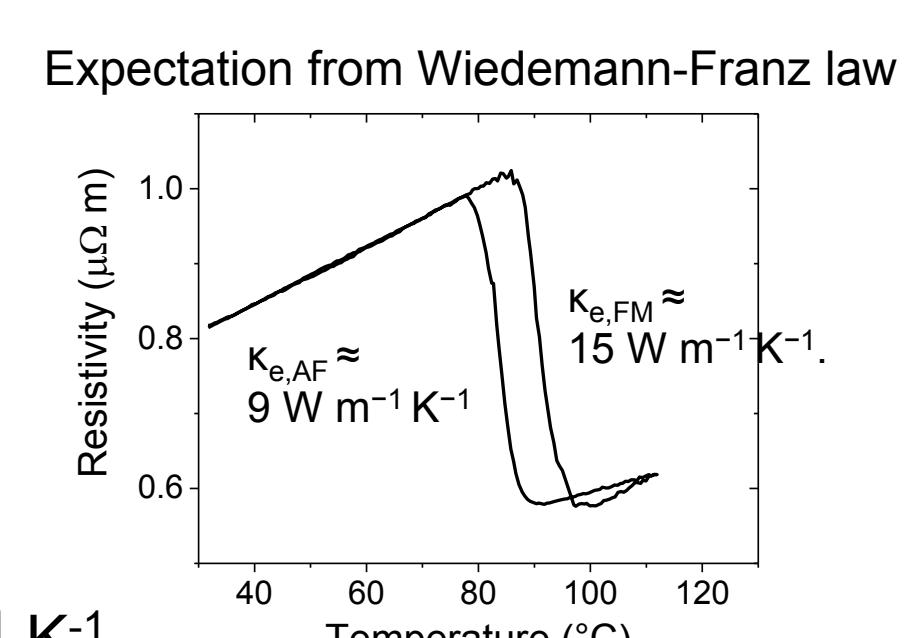
D. Fournier et al.
J. Appl. Phys., 128, 241101 (2020)

3 Thermal conductivity in AF and FM phases

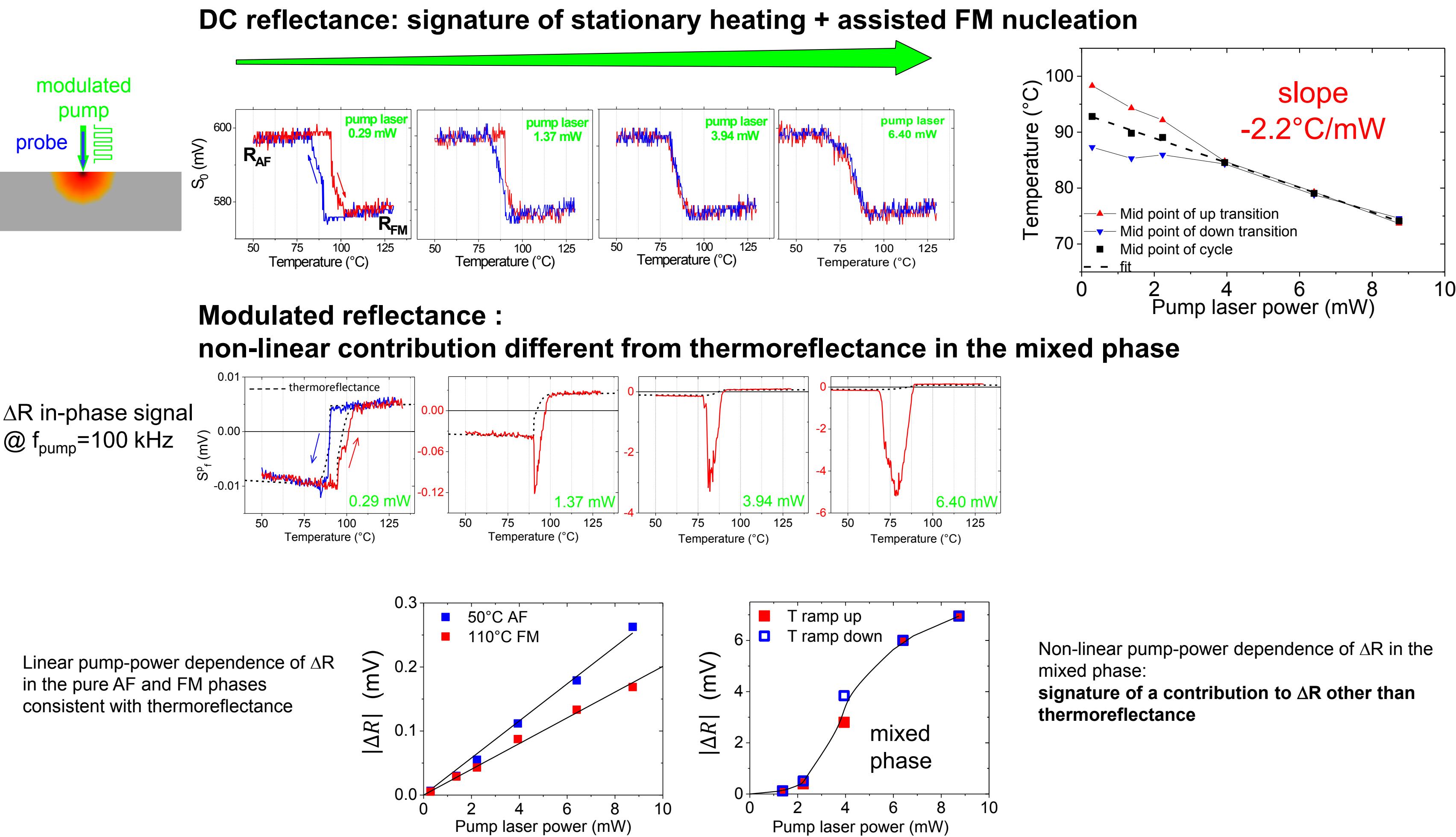


Small thermal conductivity (smaller than Fe 80 W m⁻¹ K⁻¹) larger in the AF phase than in the FM phase opposite to the expectation from Wiedemann-Franz law ($k = \sigma L T$) since the electrical conductivity $\sigma_{AF} < \sigma_{FM}$

Good agreement with theory for the AF phase (A. Castellano, M.J. Verstraete, Université de Liège)
 $k_{phonons-AF} \approx 12$, $k_{elec-AF} \approx 9$, $k_{magnons-AF} \approx 15 \rightarrow k_{total-AF} \approx 36$ W m⁻¹ K⁻¹



4 Mixed AF-FM phase: A huge contribution to the modulated reflectance other than thermoreflectance

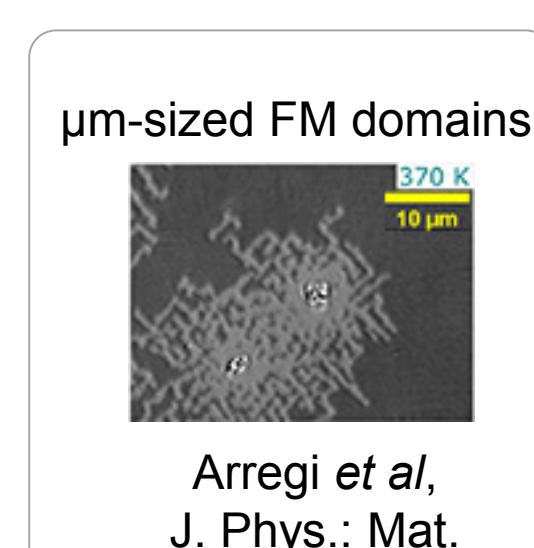


5 Pump-modulated FM fraction x

$$R = R_{AF}(1-x) + R_{FM}x$$

$$x(t) = x_0 + \Delta x(t)$$

non modulated FM fraction modulated FM fraction

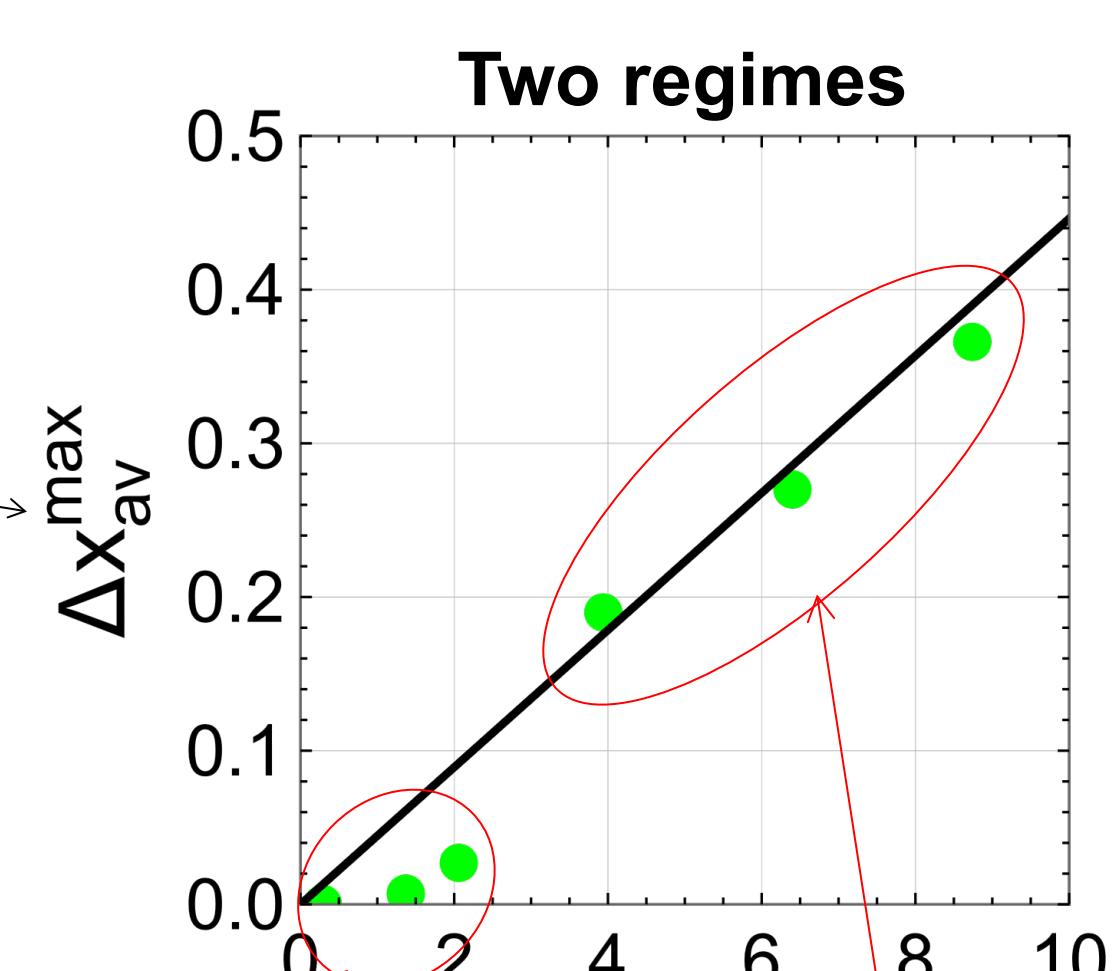
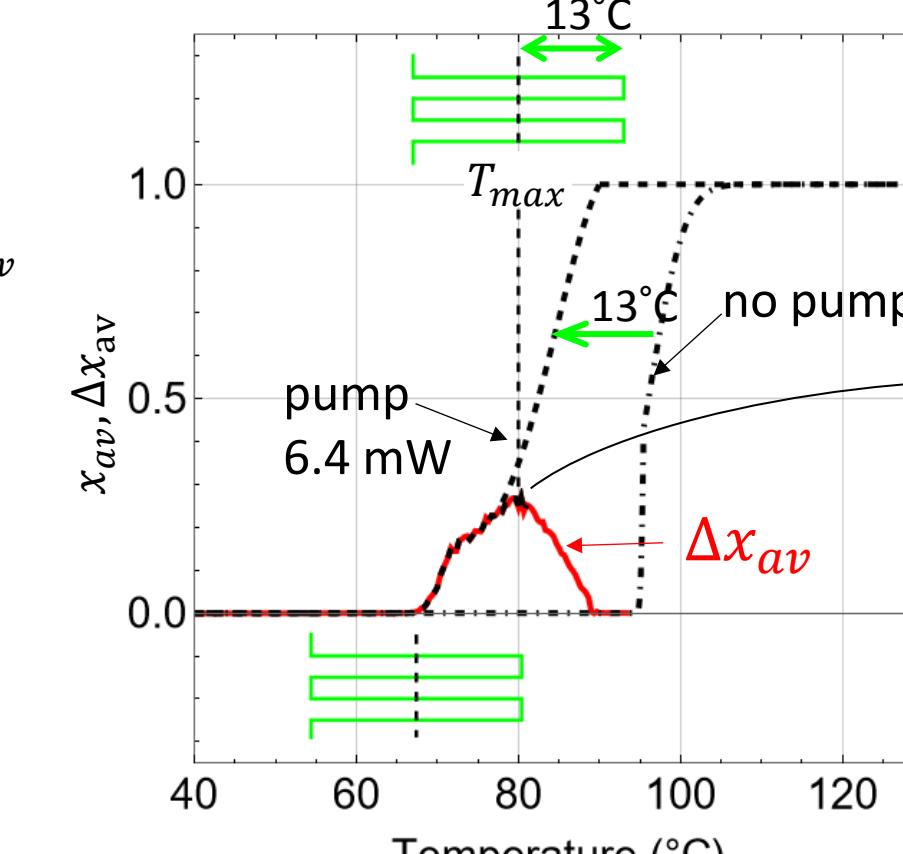
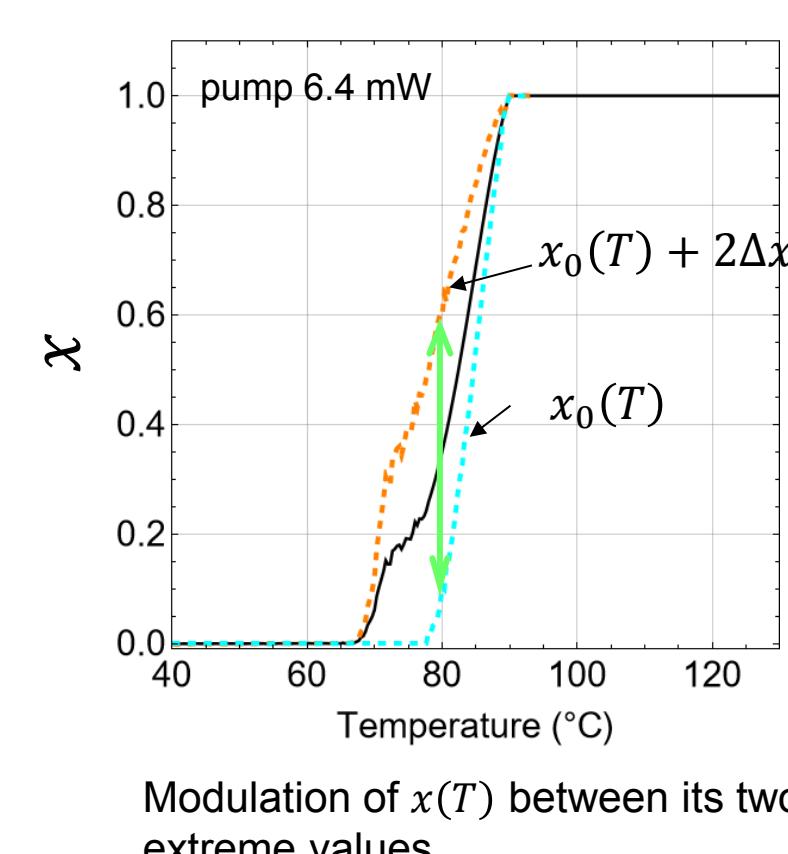
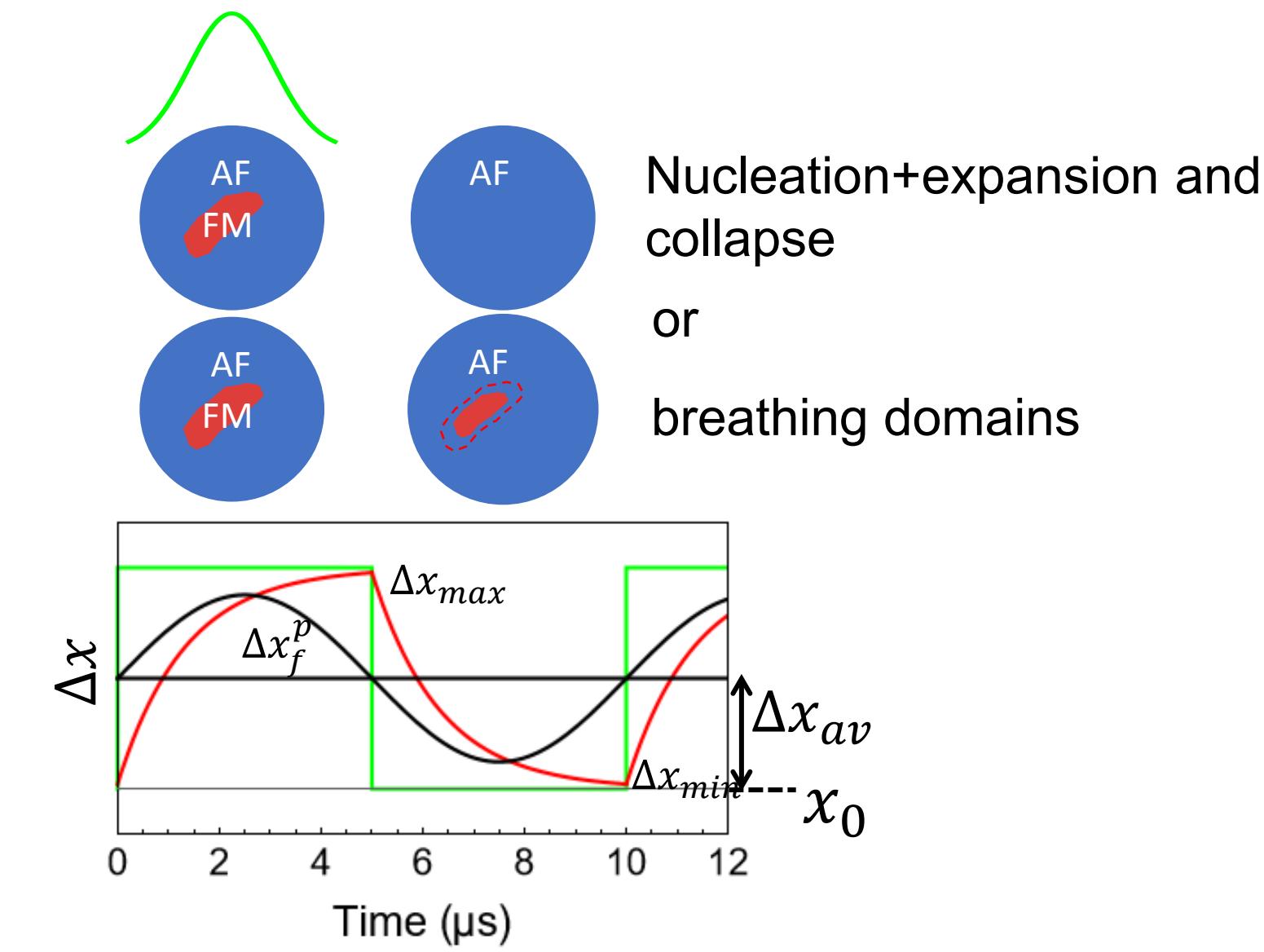


$$R = R(T_0) + \Delta R_{thermo} + \Delta R_x$$

$$\Delta R_x = \frac{dR}{dx} \Delta x = (-R_{AF} + R_{FM}) \Delta x$$

$$x_{av} = x_0 + \Delta x_{av}$$

obtained from DC reflectance obtained from in-phase and in-quadrature ΔR @ f_{pump} and DC reflectance



Conclusion

Thanks to a pretty large reflectivity difference between the AF and FM phases of FeRh (4%), a laser-induced modulation of the FM fraction can be detected in the mixed AF-FM phase. At low pump power, it likely originates from AF-FM domain wall back and forth movement and at large pump power from nucleation+expansion and collapse process across the transition. Given the risetime of the nucleation+expansion process (2-5 μs, not shown here) the domain growth process is likely governed by AF-FM domain wall velocity. The collapse is complete as long as the pump-off temperature allows to reach the fully AF phase.