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Huge laser-induced modulation of the ferromagnetic fraction in the antiferro-ferromagnetic mixed phase of FeRh

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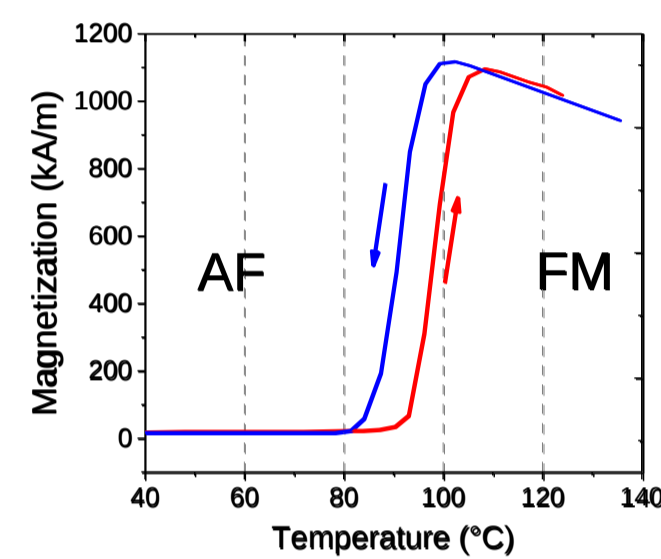
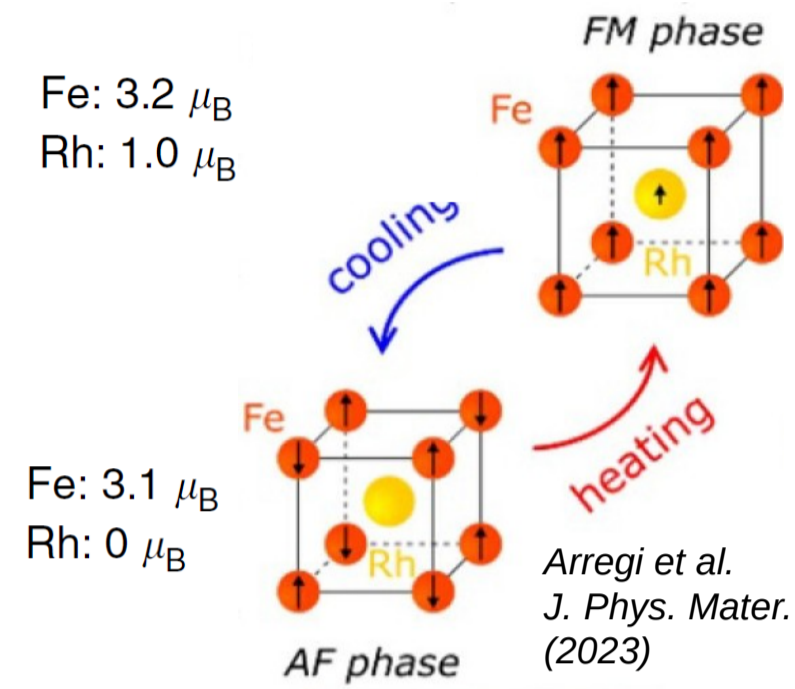
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Among magnetic materials near equi-atomic FeRh alloys, with their antiferromagnetic (AF) to ferromagnetic (FM) magneto-structural hysteretic transition near room temperature, were studied long ago in bulk form [Kouvel, *JAP* 1962, Lewis, *J. Phys.D* 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 [Martí, *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 transitions. 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 (determination of the thermal conductivity) but also allows for a quantitative determination of the power dependent laser-induced FM fraction. Moreover we show that in the mixed AF-FM phase, the system exhibits return point memory under temperature cycling, i.e. it is capable of returning back to its initial state after a minor temperature loop, despite the hysteretic nature of the transition. Surprisingly, the same distribution of nucleation sites steering the transition in quasi-static conditions can explain the thermal hysteresis in far-from-equilibrium conditions when phase change is driven by sub- μ s temperature pulses.

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
- femtosecond 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, *Matter* Arxiv 2023)

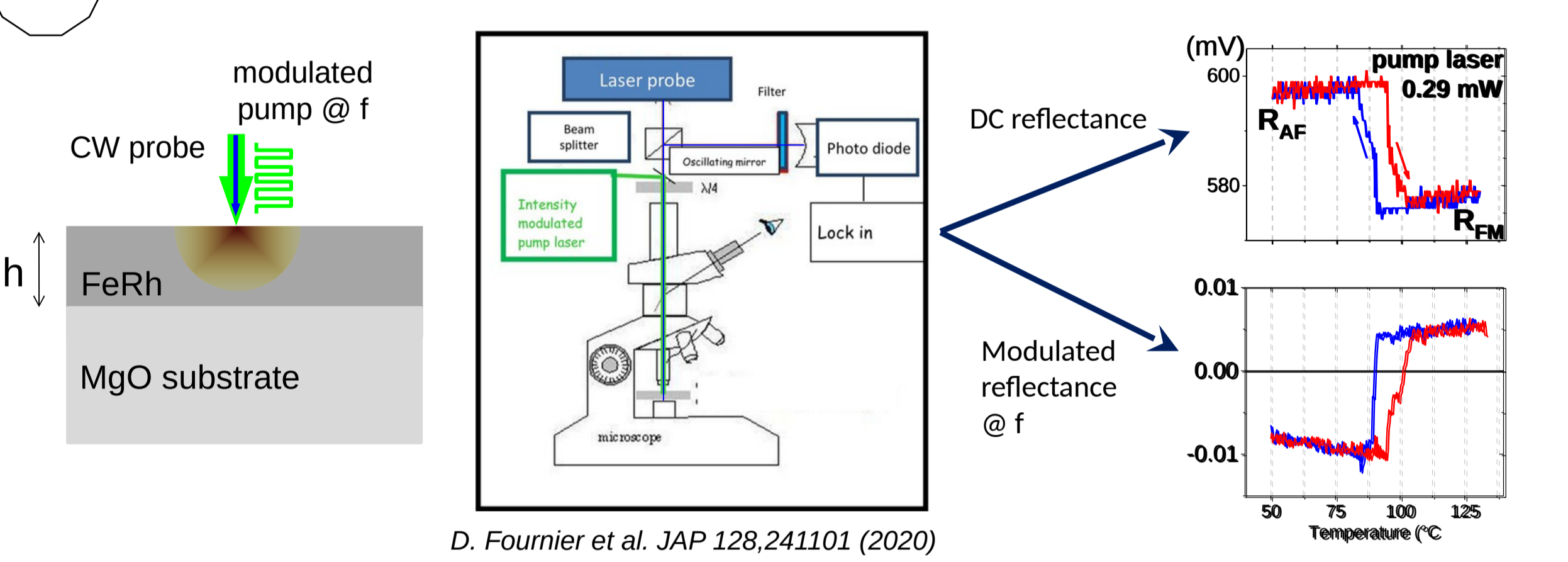


FeRh 200 nm
MgO (001) substrate

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. Uhlir et al. *Nat. Commun.* (2016)

2 Modulated reflectance: experimental setup

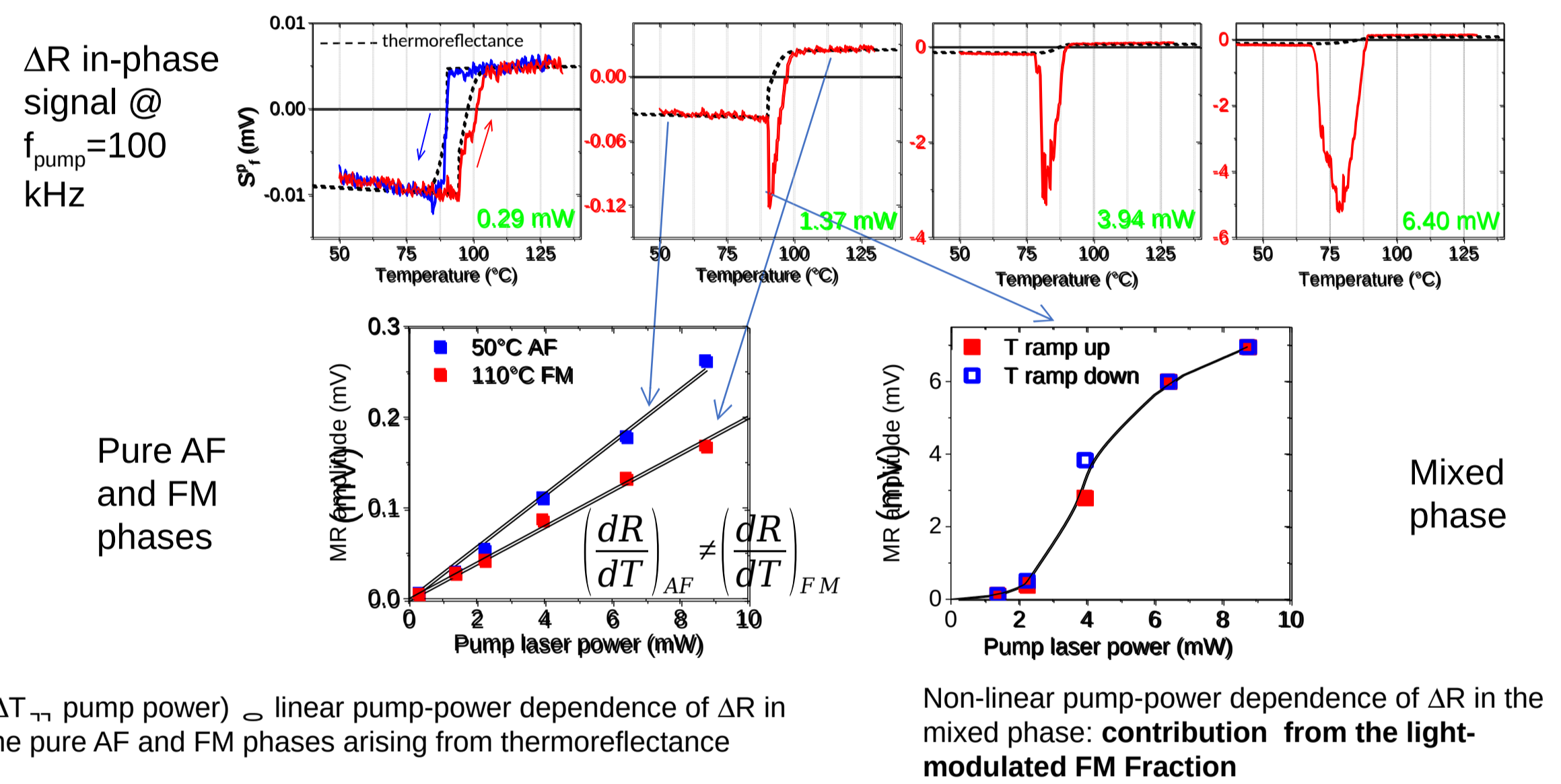


D. Fournier et al. *JAP* 128,241101 (2020)

4 Modulated reflectance: results

In AF and FM phase: only thermorefectance:

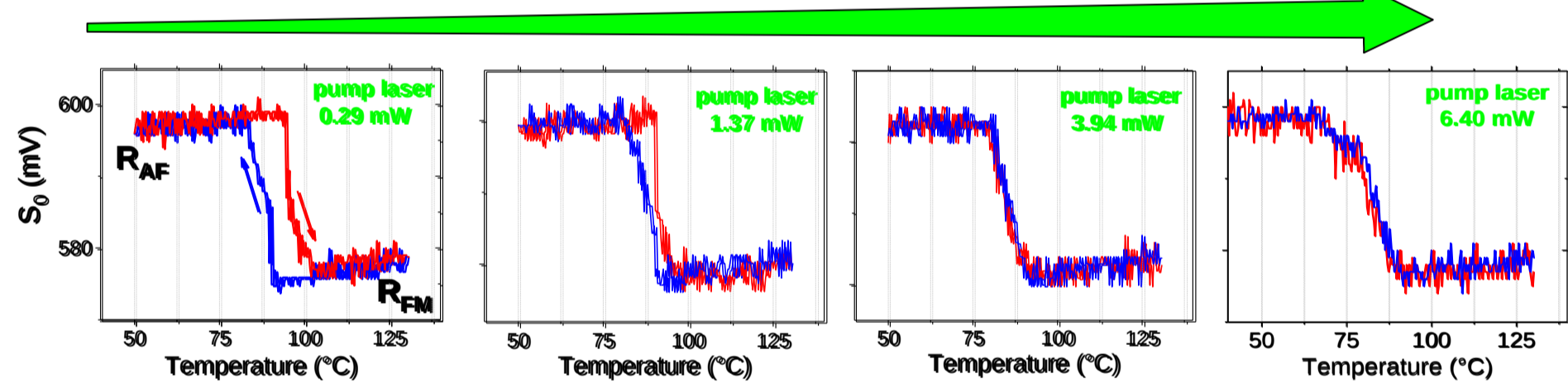
In the mixed AF-FM phase: a huge contribution from modulated FM fraction



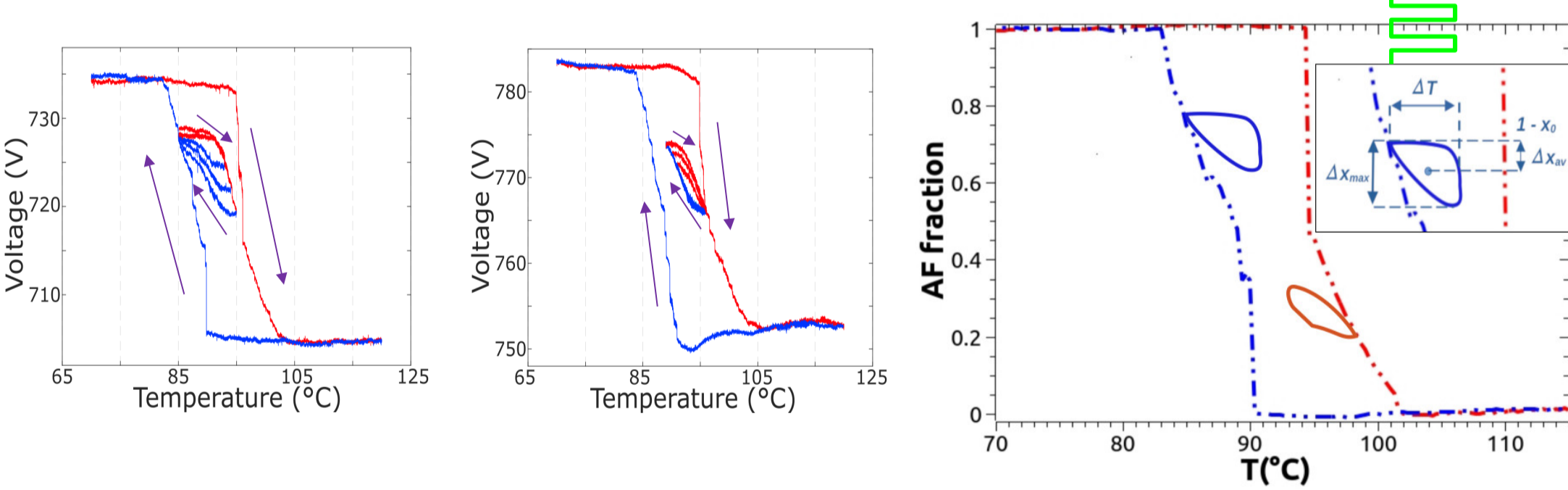
($\Delta T \rightarrow$ pump power) \rightarrow linear pump-power dependence of ΔR in the pure AF and FM phases arising from thermorefectance

Non-linear pump-power dependence of ΔR in the mixed phase: contribution from the light-modulated FM fraction

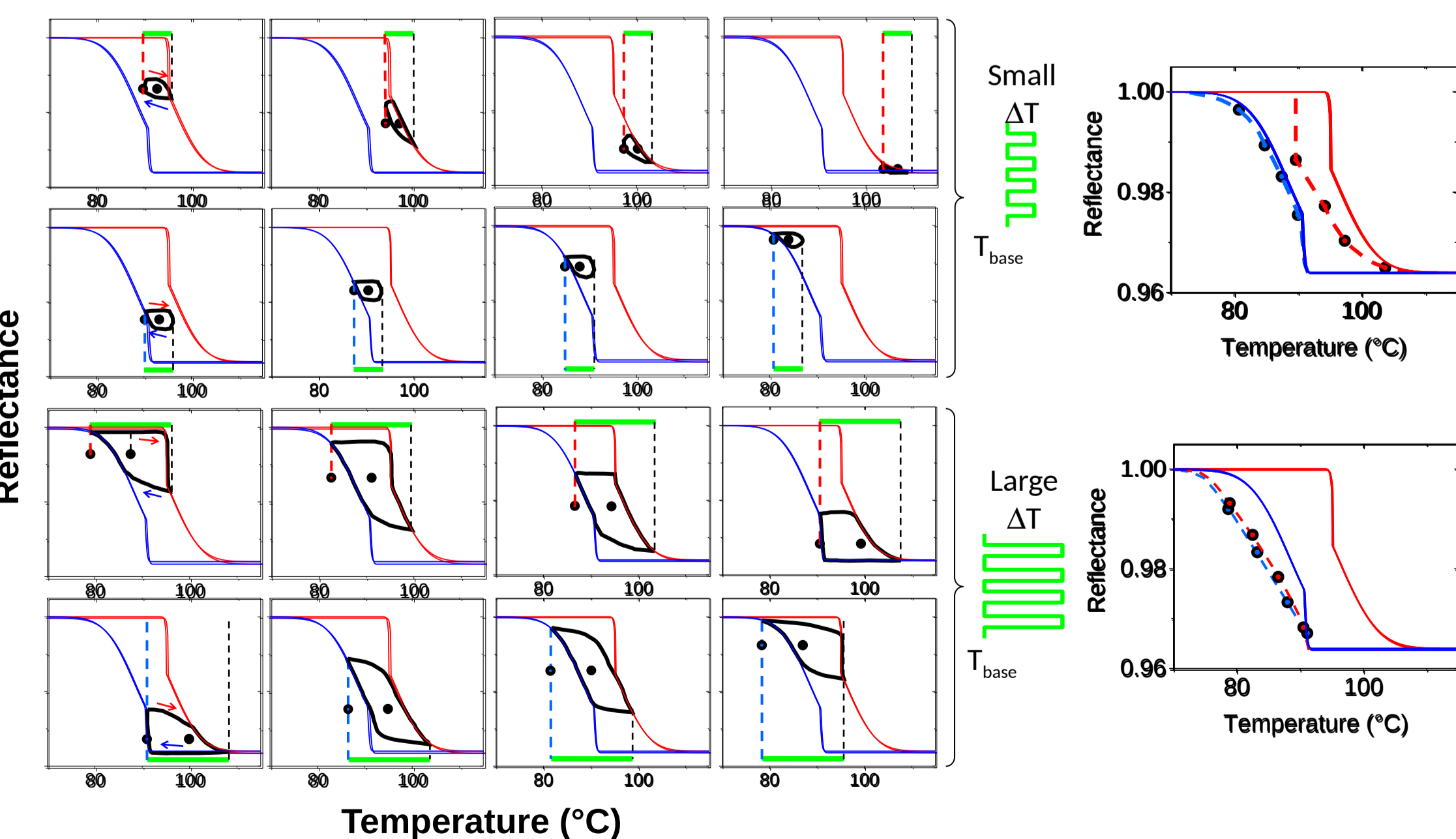
3 DC reflectance: apparent closing of the hysteresis cycle



Minor hysteresis loops: return point memory



The system exhibits **return-point memory (RPM)** a property wherein a system remembers the states at which the direction of an external driving (e.g. field, temperature...) had been reversed.



The DC signal hysteresis reduction under increasing pump power is a byproduct of plotting the DC signal as a function of T_{base} .

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5 Quantitative determination of the modulated FM fraction

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

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

non modulated FM fraction modulated FM fraction

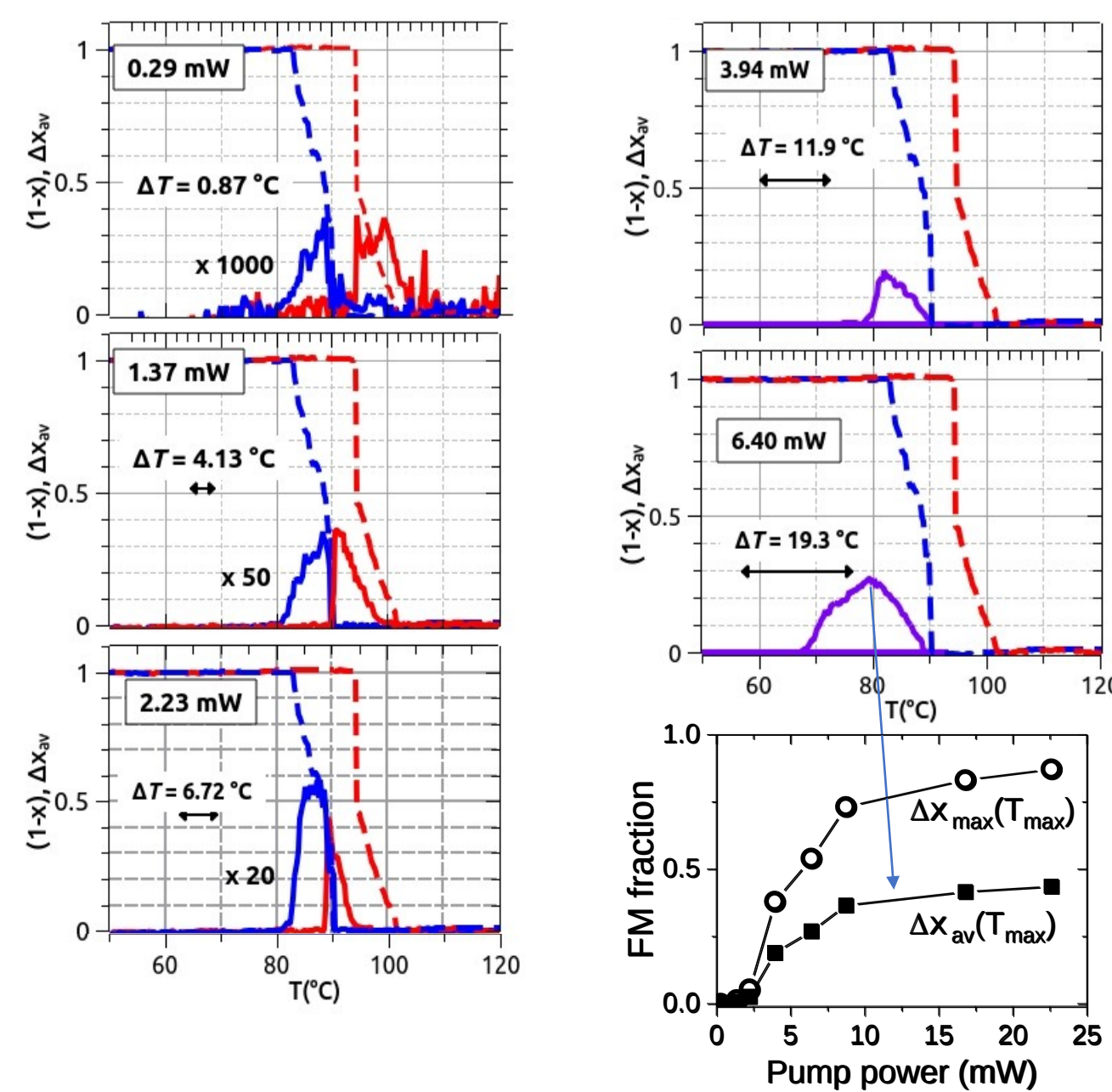
$$R = R(T_0) + \Delta R_{thermo} + \Delta R_x$$

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

The modulated FM fraction becomes detectable owing to the large reflectivity difference between the AF and FM phases and is quantitatively determined using the modulated and DC reflectance

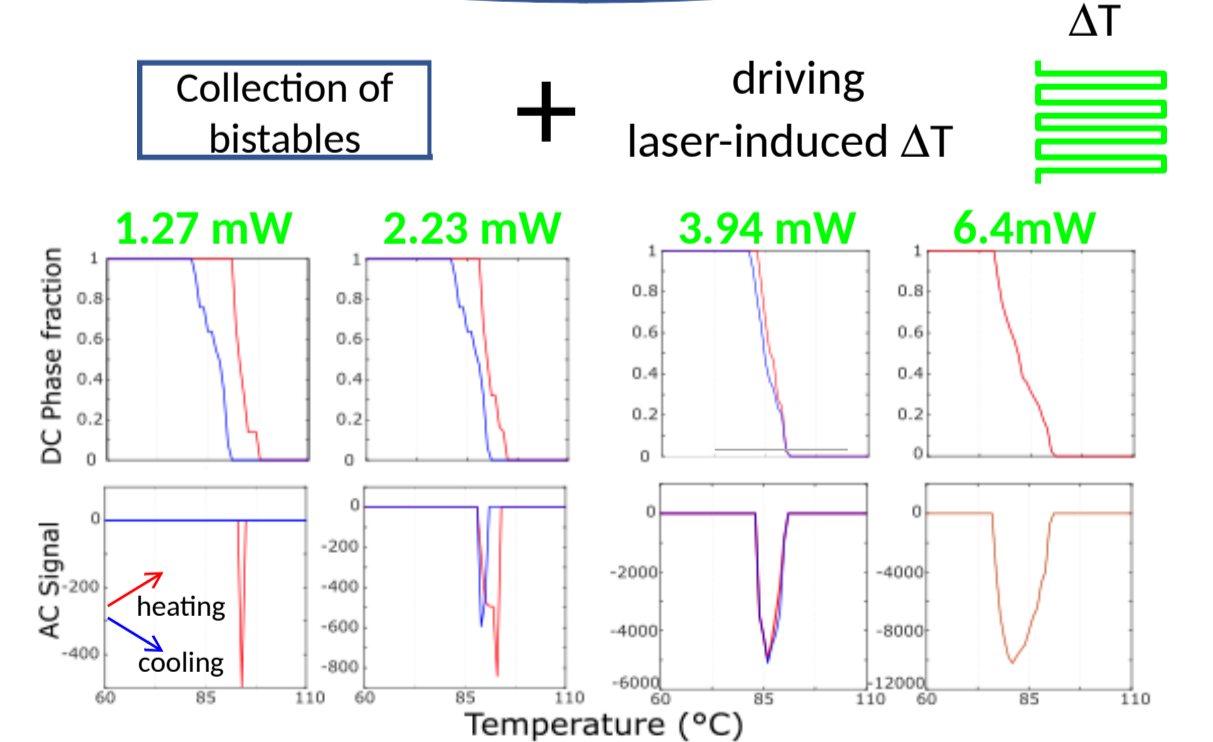
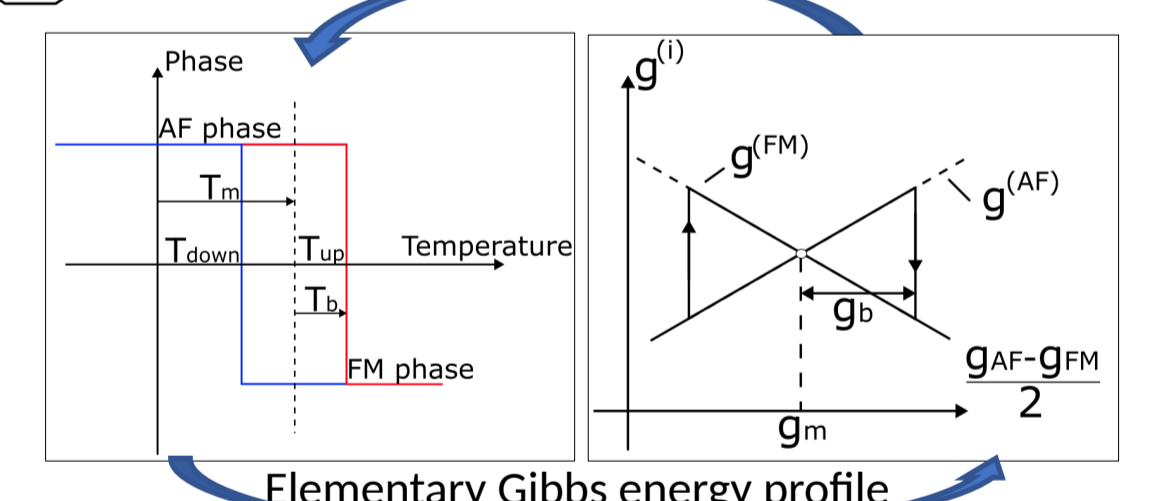
$$\Delta x_{av} = \frac{\Pi \left(1 + \frac{S_{op}^2}{S_f^2} \right)^2}{2\sqrt{2}} \frac{R_{AF}}{-R_{AF} + R_{FM}} \frac{S_f^2}{S_0^{AF}}$$

in-phase reflectance signal
out-of-phase reflectance signal
DC reflectance signal in the AF phase



At high pump power, the light modulated FM fraction reaches 90% saturation

6 Hysteresis model

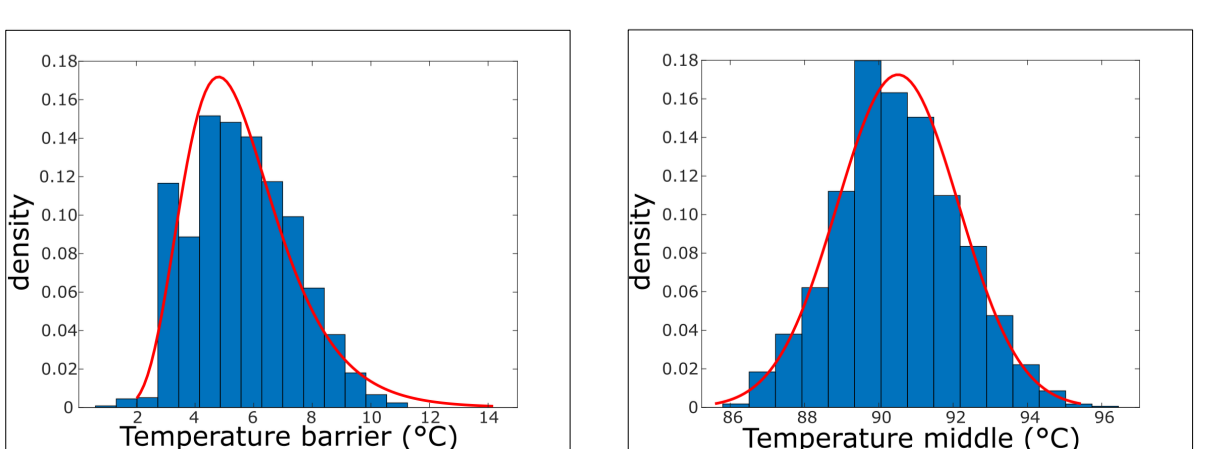


Quasi-static model can explain the apparent hysteresis closing Under periodic driving the system reaches a stationary regime dominated by its rate-independent properties.

Model Explanation

- Bistable units factorization: $p(T_m, T_b) = a \exp\left(-\frac{\ln(T_b - \mu_b)^2}{2\sigma_b^2}\right) \exp\left(-\frac{T_m^2}{2\sigma_m}\right)$
- In magnetism, this factorization is generally associated with random distribution of nucleation centers

Temperature distribution



$$T_b = \frac{T_{up} - T_{down}}{2}$$

Energy barrier

$$T_m = \frac{T_{up} + T_{down}}{2}$$

Bistable asymmetry