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Avalanche dynamics in materials for elastocaloric cooling

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Research team: Materials and Phase Transitions







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Outline

1-Introduction Structural phase transitions in solids (martensitic) Caloric effects Avalanche dynamics Experimental: Acoustic Emission (AE) & infrared (IR)

2-Recent results: AE location & dynamics of heat sources and sinks 2-1 Elastocaloric experiments 2-2 Flexocaloric experiments

3-Conclusion

Outline

1-Introduction Structural phase transitions in solids (martensitic)







Low symmetry phase exhibits symmetry related equivalent variants

Martensitic microstructure

Cu-Zn-Al single crystal, below the martensitic transition temperature

Optical image polarized light (3 mm x 2 mm)

12 equivalent martensitic variants



Metastability and non-equilibrium effects

Stress σ



Due to disorder and thermoelastic effects:

- Hysteresis
- Extended transition

Temperature T

Martensitic microstructure induced by external uniaxial stress

CuZnAl single crystal, above the martensitic transition temperature, with applied uniaxial stress in (100) direction

optical image polarized light

Two equivalent martensitic variants (twins)





Stress-strain

The martensitic transition is the key ingredient beyond interesting properties for applications:

superelasticity, high damping shape memory properties

Non-equilibrium effects showup at macroscopic scales due to different driving conditions



E.Bonnot, R.Romero, et al.,

Hysteresis in a system driven by either generalized force or displacement variables: Martensitic phase transition in single-crystalline Cu-Zn-Al Phys. Rev. B 76, 064105 (2007)

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Latent heat

Martensitic transitions proceed by nucleation and advancing fronts.

In order to advance, the fronts needs to exchange energy (latent heat)

In adiabatic conditions (fast), this results in a change of the local temperature of the sample

Front velocity

thermal conductivity





1-Introduction

Structural phase transitions in solids Caloric effects

Avalanche dynamics



Athermal FOPT and rate independent hysteresis



Competition between three time scales:

 $\tau_a \ll \tau_{dr} \ll \tau_{th}$

Influence of disorder



Order parameter



Order



The existence of disorder and the athermal behaviour of MT explains the existence of avalanche dynamics, hystheresis, and extended transitions

The characterization of this inhomogeneous and intermittent dynamics can be done with different experimental techniques

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Acoustic emission detection

C.B.Scruby. *An introduction to Acoustic Emission* J.Phys.E: Sci.Instrum. 20, 946 (1987)



Acoustic emission: hit properties

For each hit k, besides the occurrence time t_k we determine several properties, in order to characterize it:



The complex AE signal is reduced to a sequence of time instants and associated properties

 $\{(t_k; A_k, E_k, \Delta_k, \dots) \ k=1, \dots N\}$



Set of non m non linear equations with 4 unknowns (3D location)

$$t_k - t_0 = \sqrt{(x_k - x_0)^2 + (y_k - y_0)^2 + \cdots}/c$$

Simplest case: 2D location with 2 trasducers

$$X = \frac{1}{2}L\left(1 - \frac{t_2 - t_1}{t_{max}}\right)$$

$$E_0 \propto \sqrt{E_1 E_2}$$



IR measurements

Recording 2D thermal maps of the sample surface as a function of time Caution: focus, increase emissivity, avoid reflectivity,...



⊿t=0.01 s/frame, ⊿z=0.0322 mm/pixel ⊿T=0.01K





IR measurements : averaging 1D profiles

Numerical processing of 2D maps to obtain 1D temperature profiles





IR location of heat sources and sinks

Neglecting losses to air, we can consider a simple 1D Fourier equation describing the evolution of the temperature profiles

$$C\rho \frac{\partial T(x,t)}{\partial t} = \kappa \frac{\partial^2 T(x,t)}{\partial x^2} + \sigma (x,t)$$

Discretizing time $t = k \Delta t$ and space $x = i \Delta x$: T(i,k)

$$\Sigma(i,k) \equiv \frac{\sigma(i,k)}{C\rho} = \frac{1}{\Delta t} \frac{\partial T(i,k)}{\partial k} - \frac{\kappa}{C\rho\Delta x^2} \frac{\partial^2 T(i,k)}{\partial i^2}$$

Noise reduction: gaussian filter + 5 point stencil formula for the numerical derivatives 4σ

$$\overline{T}(i,k) = \sum_{n=-4\sigma} T(i,k+n)G_n(r,\sigma)$$



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Locating heat sources and sinks in tensile experiments with elastocaloric materials

Sample: Cu_{70.6}Al_{25.7}Ni_{3.7} (at%)

single crystal wire <100> direction

30mm x ϕ 1mm

Cubic \rightarrow 18R martensite

Zwick/Roell Z005 materials testing machine with displacement control 0.1 mm/s

IR Infratec 8800 camera

Two AE sensors connected to PCI2 card (Mistras).

L.Ianniciello et al., Appl. Phys. Lett. **116**, 183901 (2020) G.Capellera, et al., Appl.Phys. Lett **119**, 151905 (2021)



Stress-strain, average temperature and AE activity













In previous experiments in Cu-Zn-Al we found -1.91 but in that case the transition was not single variant.

E.Vives, D.Soto-Parra et al., Phys. Rev. B. 80, 180101R (2009)



Evolution of 1D thermal profiles with time during loading (extension)

Exothermic transition: overheating +5K Contour lines show that the heating is not homogeneous





Evolution of 1D thermal profiles with time during unloading (shrinking)

Endothermic transition: undercooling -5K





Calculation of heat sinks during unloading

Heat absorption occurs on the position of the moving austenite-martensite fronts





Dynamics of the heat sinks (I)

Position of the minimum (most negative) heat sink as a function of time





Dynamics of the heat sinks (II)

Value of the maximum cooling power (most negative Σ) as a function of time

Cooling is intermittent and there are some evidences of correlations between the two fronts Maximum heat extraction when fronts collide





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Flexocaloric experiment Unpublished (2022) Sample: Cu_{67 7}Al_{26 7}Ni_{5 6} (at%) single crystal beam: long axis <100> direction Cubic \rightarrow 18R martensite 5 mm 1 mm**‡** Top surface covered with matte paint 11 mm White dots painted in the back edge of the sample Room temperature Load cell Zwick/Roell Z005 materials testing machine with an inverted three-point bending frame IR Optical camera camera Polyestirene shield to avoid IR reflection IR Infratec 800 camera in vertical position Optical CCD camera recording the sample shape Sample Front view AE sensor AE sensors on the bending pins Sample Back view



Mechanical response:

Force versus bending amplitude during slow cycles at 1 mm/min



Hysteresis is intrinsic, related to the front movement, not related to the friction between the pins and the sample

Thermal response: bending Evolution of 1D temperature profiles and corresponding heat sources (2000 mm/min, bending amplitude $0 \rightarrow 35$ mm)





Thermal response: unbending Evolution of 1D temperature profiles and corresponding heat sinks (2000 mm/min, bending amplitude of $35 \rightarrow 0$ mm)





Acoustic response: bending + unbending

Location of AE events along the sample

1 mm/min, amplitude 25 mm. th=25dB,



Acoustic response: avalanche energy distribution (unloading)

Fit: log-normal distribution

Not a critical (powerlaw) distribution

The microstructure is highly constrained due to strain gradient and sample height



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Take home messages

Elastocaloric materials based on first-order martensitic transition show good caloric properties, due to its large latent heat

Heat absorption and release in these materials is associated with martensiteaustenite fronts movement that, at microscopic scales, follow avalanche dynamics

The heat sinks and sources are, consequently, heterogeneous in space, and move along the sample with intermittent activity

The fronts can be tracked by different experimental techniques:

-AE with location algorithms -IR imaging with inversion of Fourier equation to detect heat sources and sinks