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Influence of thermal conductivity on the dynamic response of magnetocaloric materials

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ABSTRACT

We compare the magnetocaloric effect of samples prepared with different thermal conductivities to investigate the potential of composite materials. By applying the magnetic field under operating conditions we test the material's response and compare this to heat transfer simulations in order to check the reliability of the adiabatic temperature change probe used. As a result of this study we highlight how the material's thermal conductivity influences τ, the time constant of temperature change. This parameter ultimately limits the maximum frequency of a refrigerant cycle and offers fundamental information about the correlation between thermal conductivity and the magnetocaloric effect.

Influence de la conductivité thermique sur la réponse dynamique des matériaux magnétocaloriques

Mots clés : Transitions magnétiques de premier ordre ; Effet magnétocalorique ; Conductivité thermique ; Constante de temps

1. Introduction

Minimization of energy consumption is currently one of the most demanding research challenges, motivating the development of new technical solutions to slow down climate change and the depletion of available resources. As a large amount of worldwide energy consumption can be attributed to cooling devices (Gutfleisch et al., 2011; Sandeman, 2011), the development of a new generation of more efficient and environmentally

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Nomenclature

T	absolute temperature [K]
t	time [s]
H	magnetic field [A m^{-1}]
m	mass [kg]
c_p	specific heat capacity [J kg^{-1} K^{-1}]
k	thermal conductivity [W m^{-1} K^{-1}]
T_C	Curie temperature [K]
$\dot{q}$	heat source [W m^{-3}]
C	electric capacity [F]
Q	electric charge [C]
I	electric current [A]
R	electric resistance [Ω]
Δs_T	specific isothermal entropy change [J kg^{-1} K^{-1}]
ΔT_{ad}	adiabatic temperature change [K]
ΔT_{hyst}	temperature hysteresis [K]
Δx	spatial step of the simulation mesh [μm]
Δt	time step of the simulation mesh [μs]
Δt_{field}	magnetic field rise time [s]
τ	adiabatic temperature change time constant [s]
$\mu 0$	permeability of vacuum [$4\pi 10^{-7}$ H m^{-1}]
ρ	density [kg m^{-3}]

friendly refrigerators would therefore constitute a technological revolution.

Solid-state magnetic cooling is one such promising technology that has the potential to realize new, more efficient cooling devices without the need to use hydrofluorocarbon (HFC) greenhouse gases. The magnetocaloric effect (MCE), which constitutes the working principle of these machines, is an entropy (Δs_T) or a temperature (ΔT_{ad}) change induced by a changing magnetic field under isothermal or adiabatic conditions, respectively (Pecharsky et al., 2001; Smith et al., 2012; Tishin and Spichkin, 2003). The Maxwell relations, which are commonly used to quantify this effect (Brück, 2005), show that the MCE is maximum in the neighborhood of magnetic phase transitions. Back in 1997 the discovery of a giant magnetocaloric effect (GMCE) in $Gd_5(Si,Ge)_4$ compounds highlighted that large Δs_T values could be generated under an external field due to the contribution of latent heat (in this case originating from a structural phase change) (Pecharsky and Gschneidner, 1997). This idea is arguably the platform on which the current magnetocaloric community was built, and many other interesting first order materials have since been discovered (Hu et al., 2001; Krenke et al., 2005; Sandeman, 2012; Tegus et al., 2002; Trung et al., 2010; Wada and Tanabe, 2001).

The processes studied so far are generally solid-state phase transitions characterized by a specific latent heat of one to two orders of magnitude lower than the specific latent heats of vaporization seen in the HFC fluids. In order to develop a competitive device that pumps the same amount of heat as a commercial refrigerator while achieving a higher efficiency (for a similar mass of refrigerant), magnetocaloric materials would have to sustain thermodynamic cycles of a few Hertz compared to 0.03 Hz for small (1–1.5 kW cooling power) vapor compression refrigerators.[1] With this requirement in mind, the study of the material's response under prototypical operating conditions becomes increasingly important (Bahl et al., 2008; Kuz'min, 2007; Legait et al., 2014; Lyubina, 2011; Lyubina et al., 2010; Moore et al., 2009, 2013; Nielsen and Engelbrecht, 2012). An experimental characterization technique that is able to simulate real working conditions will therefore offer invaluable information about the reproducibility and dynamic response of a magnetocaloric material (Franco et al., 2012). The interplay between the material's thermal conductivity and the dynamic response of the magnetocaloric material to the applied magnetic field will determine the maximum achievable operation frequency (Lovell et al., 2014). In addition, magnetic field-induced structural fatigue is another feature that will need to be considered when engineering future materials.

Many of the most promising systems studied show first order phase transitions that are irreversible if exposed to the magnetic field changes typically achievable by permanent magnets. This self-limiting issue has motivated many groups to look for different controlling parameters such as electric field, pressure and stress in order to more easily drive these solid–solid phase transitions (Czernuszewicz et al., 2014; Fähler et al., 2012; Mañosa et al., 2010, 2013; Moya et al., 2013, 2014; Ozbolt et al., 2014). However, any future energy-conversion device based on one of these caloric effects will also have to operate high frequency thermodynamic cycles. The development of good working systems will therefore follow a similar path to that already followed for magnetocaloric materials. In other words, studies on the relationship between the material's thermal conductivity and electro-, baro-, elasto-caloric effects together with investigation of the evolution of structural fatigue induced by the ongoing application of electric field or pressure or stress will be needed.

In this work we study the dynamic response to the external field of magnetocaloric materials prepared with different thermal conductivities. We start by presenting the outcomes of two $La_{0.67}Ca_{0.33}MnO_3$ manganite samples. Our initial observations indicate that the shape of the thermodynamic cycles performed in these two cases is markedly different. To understand the origin of this behavior we perform heat transfer simulations and repeat the same characterization for a gadolinium plate and two MnFePSi-based alloys, also engineered to have different thermal conductivities.

2. Experimental

$La_{0.67}Ca_{0.33}MnO_3$ (labeled LCMO) pellets were prepared from polycrystalline powders synthesized by the glycine nitrate process, as presented by Turcaud et al. (2013). This material, beyond its complex and fascinating behavior across the metal–insulator transition (Turcaud et al., 2014), is interesting also for energy conversion applications since recently similar compositions showed promising results when tested in a refrigeration test device with the combination of a modest and reversible MCE

[1]Technical data extracted from BITZER Software v6.4.3 rev.1353 and provided by Dr. M. Mazzani, researcher at Zanotti S.p.a. (private communication).