

Relaxation effects in magnetic-field-induced martensitic transformation of an Ni–Mn–In–Co alloy

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Received 25 October 2013; received in revised form 26 February 2014; accepted 28 February 2014

Available online 31 March 2014

Abstract

The effect of isothermal stepwise changes of an applied magnetic field in an Ni–Mn–In–Co metamagnetic shape memory alloy has been studied. It has been found that relaxations towards equilibrium states occur under isothermal and isofield conditions, in both direct and reverse martensitic transformations (upon decreasing or increasing field sequences, respectively). In all cases, the relaxation follows a logarithmic time dependence, which can be monitored through the time dependence of magnetization $M(t)$. The logarithmic dependence brings close analogies with the thermal fluctuation magnetic after-effect, associated with the thermally assisted rearrangement of magnetic domains after a sudden variation of the magnetic field. Following this analogy, it has been found that the viscosity coefficient Z (slope of $M(t)$ vs. $\ln t$) is directly proportional to the transformation rate in a continuous cycle. This result generalizes what has been found in temperature-induced transformations, although in field-induced transformations the proportionality is temperature dependent. This result also allows for an understanding of the existence of isothermal–isofield “windows” where the relaxations can take place, and the experimental results found in quite different systems can be easily rationalized in this way. Finally, the features of pseudo-transformation–temperature–time diagrams in this alloy can be rationalized according to the proposed dependence of Z .

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Keywords: Phase transformation kinetics; Metamagnetism; Martensitic phase transformation; Magnetic shape memory alloys

1. Introduction

Ferromagnetic shape memory alloys (FSMAs) are a class of multiferroic materials which are currently generating a great deal of interest due to their unique properties, such as their ability to undergo giant magnetic-field-induced strains by variant reorientation, as in Ni–Mn–Ga [1], or associated with a magnetic-field-induced (reverse) transformation – a magnetic shape memory effect (MSM) – which take place in Ni–Mn–X type alloys (typically $X = \text{In, Sn, Sb}$) [2]. In these systems, a large inverse magnetocaloric effect has also been found [3,4]. Both this effect

and the MSM effect are related to the martensitic transformation (MT) accompanied by a large change in magnetic order between both phases.

Even though it is commonly accepted that MTs have an athermal character, some cases of isothermal behaviour have been known for several years. For example, in some Fe–Ni–Mn systems, the application of high magnetic fields changes the originally isothermal kinetics to athermal [5]; in Cu–Al–Ni the MT occurs after some incubation period during isothermal holding at a temperature higher than the martensitic start temperature, T_{Ms} , and similarly, the reverse MT takes place upon isothermal holding at $T < T_{As}$ [6]. Isothermal features are also present in the magnetostructural transition shown by Ni–Mn–X metamagnetic shape memory alloys [7–11]. Moreover, the effect of the

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applied magnetic field seems to be very important in relation to the isothermal characteristics, showing some analogies with the above-mentioned Fe–Ni–Mn system, although it has to be kept in mind that the magnetic field favours the appearance of the martensitic phase in the Fe-based alloy, contrary to what occurs in Ni–Mn–X alloys. It has been recently reported that in Ni–Mn–X alloys the magnetic field leads to the arrest of the MT by stabilizing the parent (high magnetization) phase [7,8,12,13] and it is in the arrested MT where the isothermal behaviour is easily encountered. Nevertheless, Kustov et al. showed in an Ni–Mn–In–Co alloy that the arrested state is not a necessary condition for the isothermal behaviour [10]. In this sense, it is worth mentioning that very recently, isothermal MTs were found in Ni–Ti alloys, where no external field is applied; thus the isothermal behaviour does not have a necessary relation with any externally produced arrest of the transformation [14].

The progress of the isothermal transformation in Ni–Mn–X and Ni–Ti alloys which can be found by stopping the cooling/heating cycle within the respective transformation range, and/or by changing the magnetic field in the case of metamagnetic shape memory alloys, can be properly described by a logarithmic law, as has been shown in Refs. [10,14–17]. The logarithmic dependence brings close analogies with the thermal fluctuation magnetic after-effect, associated with the thermally assisted rearrangement of magnetic domains after a sudden variation of the magnetic field. It is considered that the overall metastable behaviour of the system, related to a complicated free energy surface landscape with a large number of local minima, can be decomposed into a superposition of many bistable contributions relaxing over a set of energy barriers [18,10]. In the case of relaxations in MT, the physical mechanism is different (as we are not dealing with pure rearrangements of magnetic domains), but the presence of different energy barriers through which the direct/reverse transformation takes place can be considered as an appropriate picture.

From these previous experimental data it has been noticed that there is an interplay between the temperature and the magnetic field in the kinetics of the isothermal transformation through the slope of the logarithmic time dependence, Z [15,16]. In addition, some regularities have been observed in the relation between the martensite fraction present during the isothermal holding and the parameter Z , which could be ascribed to the role of martensite–austenite interfaces in the kinetics of the isothermal transformation [16].

Any possible application of shape memory alloys, whether ferromagnetic or not, often relies on the properties of an athermal MT; thus to clarify, the conditions which control the thermal/athermal kinetics are of obvious interest. Therefore, progress in the understanding of the isothermal/athermal character of the MTs in general, not only in the metamagnetic alloys, is necessary.

The aim of this paper is to analyse in a more systematic way the isothermal transformation features produced by

isothermal (stepwise) changes of the applied magnetic field in an Ni–Mn–In–Co ferromagnetic shape memory alloy, at temperatures close or below the martensitic finish temperature, T_{Mf} , in such a way that sequences of increasing or decreasing applied magnetic field will promote the reverse or direct (isothermal) MT, respectively. These data are compared with the isothermal characteristics, obtained in the same alloy, by stopping cooling–heating temperature-induced transformation cycles under a constant field. In the case of temperature-induced cycles, proportionality was found between the isothermal transformation rate, Z , and the transformation rate in a continuous cycle [10]. This relation is discussed and extended to isothermal, magnetic-field-induced MT.

2. Experimental procedure

An $\text{Ni}_{45.0}\text{Mn}_{36.7}\text{In}_{13.3}\text{Co}_{5.0}$ (nominal composition, at.%) alloy produced by arc-melting, with several consecutive remeltings, has been used. Samples for magnetic measurements of parallelepiped shape of $\sim 0.5 \times 1 \times 4 \text{ mm}^3$ were encapsulated in vacuum-sealed quartz tubes and further homogenized by 24 h annealing at 1170 K followed by water-quenching. After this treatment the alloy composition, as obtained by energy-dispersive X-ray spectroscopy, was $\text{Ni}_{45.5}\text{Mn}_{35.3}\text{In}_{12.7}\text{Co}_{6.5}$. It is known that the MT in these alloys can be shifted to lower temperatures by increasing the degree of $L2_1$ order [19] and that in this case the arrest of the transformation produced by the magnetic field is much more intense than in samples with a lower degree of order [10]. Therefore, the samples were subjected to an additional treatment of 15 min at 1070 K followed by slow cooling (hereafter referred to as SC samples – this naming has been chosen in order to be coherent with equivalent treatment in other papers). In this case T_{Ms} was close to 260 K and the structure of martensite was a mixture of seven-layered (14 M) and no-modulated (2 M) [9]. Absence of decomposition and/or precipitation was confirmed by optical and transmission electron microscopy. A Squid magnetometer [20] was used to measure the magnetization in series at constant temperature, under different applied fields, in the range 1–7 T. The time dependence of the magnetization $M(t)$, which indicates the progress of the direct/reverse transformation, is the physical magnitude from which the kinetics of the isothermal transformation is obtained. Once the temperature and the field are stable, $M(t)$ follows a logarithmic law, in analogy with the results obtained by AC and DC resistance measurements [10]:

$$M(t) = M_0 + Z \ln(1 + t/t_0) \quad (1)$$

where $M_0 = M(t = 0)$ and t_0 , Z and M_0 are the three parameters fitting the experimental data to the dependence given by Eq. (1). t_0 takes into account possible transient effects and is related to the time required for the Squid to reach a constant value of the magnetic field after its variation. Following the analogy with the thermal fluctuation magnetic after-effect, Z can be considered equivalent to

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