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Modeling the fractional magnetic states of magnetostructural transformations

Edward Della Torre [a], Hatem ElBidweihy [a,*], Virgil Provenzano [b], Lawrence H. Bennett [a]

[a] Department of Electrical and Computer Engineering, The George Washington University, Washington, DC 20052, USA
[b] National Institute for Standards and Technology, Gaithersburg, MD 20899, USA

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ABSTRACT

The large inverse magnetocaloric effect (MCE) in the off-stoichiometric Heusler alloys occurs at a critical temperature near room temperature. At this temperature, the material is in a mixed-state and can have a variable ratio of two stable magnetic crystallographic-states; a high magnetization state (HM) and a low magnetization state (LM). The field-induced thermal hysteresis in the virgin curve of $Ni_{50}Mn_{35}In_{15}$ and the virgin first-order reversal curves (VFORC) are presented. A model is introduced to describe the descending branches of these curves based on the different magnetic fields of conversion (from HM to LM). Using limited measurements, the model is used as a tool to determine the fractions of the two crystallographic-states within the mixed-state region.

1. Introduction

In the search for materials with giant magnetocaloric effect (MCE), attention has been focused recently on certain Heusler alloys. These materials have a large inverse MCE near room temperature and therefore may be useful for magnetic refrigeration applications.This effect in certain Heusler alloys occurs at a critical temperature. We believe that this occurs at the Kittel temperature [1] by a critical change in the lattice parameter where the sign of the exchange energy reverses. Alternately, it has been suggested that it is due to a martensite–austenite transition at this temperature. In either case, the mechanism is not germane to this model. At the critical temperature, the material is in a mixed-state where two distinct stable magnetic crystallographic-states are present; a high magnetization state (HM) and a low magnetization state (LM) [2]. A mixed-state refers to the presence of two distinct crystal structures, but only their magnet structures are measured and discussed in this paper. Experimental analysis has been used to determine the fractions of these crystallographic states, and their effect on the hysteretic behavior of Ni–Mn-based Heusler alloys [3,4]. Differently, we present a magnetization model which can be used as a tool for analyzing the magnetic behavior and for determining the fractions of the crystallographic-states in the mixed-state region.

2. Material and method

The $Ni_{50}Mn_{35}In_{15}$ sample was prepared by arc melting appropriate amounts of the component elements, using a water-cooled copper hearth in an argon atmosphere under ambient pressure. The sample was then homogenized for 2 h at 800 °C in an evacuated quartz tube and then quenched in ice water. The sample's structure and magnetic properties are characterized by X-ray diffraction spectroscopy and SQUID magnetometry, respectively.

Our data [2,5] show that the first-order transition occurs within the 270–290 K temperature range. All the data presented in this paper were measured at the critical temperature (280 K), where the alloy is in a mixed crystallographic-state. Two virgin magnetization curves measured at 280 K are shown in Fig. 1. The field is cycled between zero and saturation. It is seen that the ascending curve of the demagnetized specimen that approached the critical temperature (280 K) from below (from 250 K) lies well below that of the demagnetized specimen that approached the critical temperature from above (from 300 K). The descending branches from saturation to zero field are identical for both cases. We conclude that there is an irreversible process of thermal hysteresis in the virgin curve. Thermal hysteresis occurs at this temperature between the two distinct crystallographic-states; HM and LM. The transformation from one state to the other is asymmetrical and can be field-induced in one direction only. Under the same conditions, the material can be in different crystallographic-states at the same temperature depending upon whether it was cooled down or heated up to that temperature. When the LM crystallographic-state transforms to the HM one, the exchange energy changes its sign. This near-neighbor exchange sign reversal is the principal loss mechanism in these materials.

* Corresponding author.
E-mail address: hatem@gwmail.gwu.edu (H. ElBidweihy).

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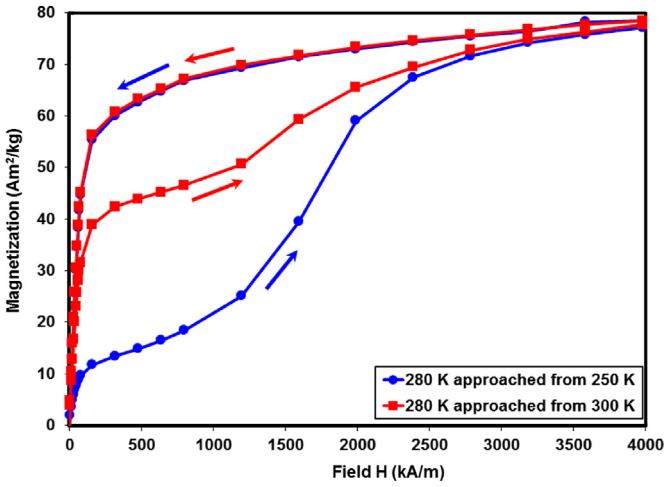

Fig. 1. *M* vs. *H* virgin curves at 280 K approached from 250 K (blue circles) and 300 K (red squares). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

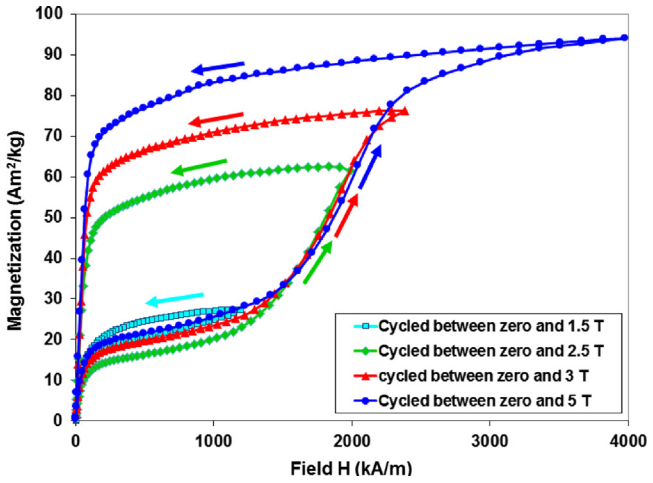

Fig. 2. *M* vs. *H* at 280 K for four different maximum values of the applied field.

The material is initially demagnetized by applying a temperature above the Curie temperature. When the temperature is lowered to the critical temperature, the majority of the spins will be in the HM crystallographic-state. If the temperature is lowered well below the critical temperature and then raised to the critical temperature, the majority of the spins will be in the LM crystallographic-state. This can be tested by applying an increasing magnetic field to the two cases. It is seen that the crystallographic-state that has a majority of HM coupled spins will sill start at a higher magnetization and have a smaller number of magnetic conversions than the crystallographic-state that has a majority of LM coupled spins. However, in high fields they will both achieve the same magnetization and will behave the same afterwards. The virgin magnetization curves for four different maximum values of the applied field are shown in Fig. 2.

The virgin curve and the loop measured second at 280 K for an applied field of 5 T are shown in Fig. 3. Upon decreasing the applied field below a certain level, h_d, a certain fraction of the spins in the HM crystallographic-state relaxes back to the LM crystallographic-state. We will assume that the fraction relaxing is directly proportional to difference in magnetization between the HM and the LM crystallographic-states. Then, there is no relaxation when the material is in the LM crystallographic-state. There is no further relaxation when the applied field falls below 100 kA/m.

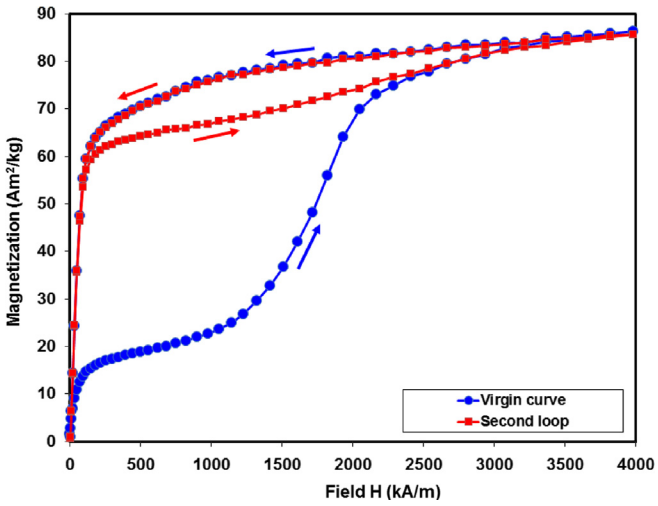

Fig. 3. *M* vs. *H* at 280 K for a virgin curve and a loop measured second.

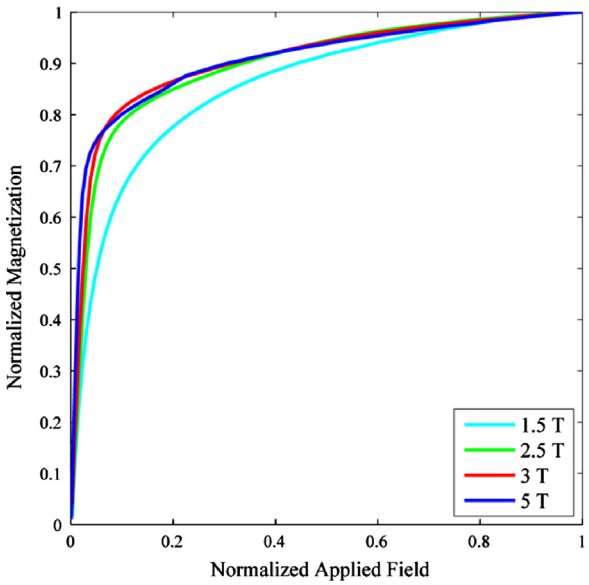

Fig. 4. The normalized curves of the four descending curves in Fig. 2.

On cycling the applied field, a hysteresis loop forms both of which critical fields are of the same sign.

3. Discussion

Normalizing the four descending branches in Fig. 2 each to their maximum magnetization and their maximum field reached gives the curves in Fig. 4. The normalized curves have essentially the same shape indicating that they all come from a similar process. The normalized curve of the lowest applied field (1.5 T) is slightly different because it does not have as much coupled spins in the HM crystallographic-state as those induced by the higher fields.

The normalization factor is plotted as a function of the maximum value of the applied field reached in Fig. 5. This factor is the ratio of the saturation magnetization of each of the four curves (Fig. 2) to the saturation magnetization of the 5 T curve. The obtained values are 0.85, 0.7 and 0.3 for the 3, 2.5 and 1.5 T curves, respectively. If the critical fields are normally distributed, the cumulative distribution should be an error function. Fitting the