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History-dependent nucleation and growth of the martensitic phase in the magnetic shape memory alloy $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$

A. Banerjee, P. Chaddah, S. Dash, Kranti Kumar, and Archana Lakhani

UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore 452001, Madhya Pradesh, India

X. Chen and R. V. Ramanujan

School of Materials Science and Engineering, Nanyang Technological University, N4.1-01-18, 50 Nanyang Avenue, Singapore 639798

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We study through the time evolution of magnetization the low-temperature (T) dynamics of the metastable coexisting phases created by traversing different paths in magnetic field H and T space in a shape memory alloy system, $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$. It is shown that these coexisting phases consisting of a fraction of kinetically arrested austenite phase and a remaining fraction of low- T equilibrium martensitic phase undergo a slow relaxation to low magnetization (martensitic) state but with very different thermomagnetic history-dependent rates at the same T and H . We discovered that, when the nucleation of the martensitic phase is initiated at much lower T through the de-arrest of the glasslike arrested state contrasted with the respective first-order transformation (through supercooling at much higher T), the long-time relaxation rate scales with the nonequilibrium phase fraction but has a very weak dependence on T . This is explained on the basis of the H - T path dependent size of the critical radii of the nuclei and the subsequent growth of the equilibrium phase through the motion of the interface.

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I. INTRODUCTION

The intriguing process of nucleation and growth of competing magnetic order in the course of first-order magnetic transition gives rise to interesting physical phenomena in a variety of materials of current interest.¹⁻³ There have been many reports in materials ranging across intermetallics,⁴⁻⁸ CMR manganites,⁸⁻¹³ and multiferroics¹⁴ of the arrest of first-order transformation kinetics before its completion. Further transformation to the low-temperature equilibrium phase does not take place down to lowest temperature (T) indicating that the process of nucleation and growth is inhibited. Thus, at low temperatures, a fraction of transformed low-temperature equilibrium phase coexists with the remaining fraction of kinetically arrested (KA) high-temperature phase that shows non-ergodic behavior and glasslike dynamic response. The effect of KA has been recently reported extensively in magnetic shape memory (MSM) materials across their first-order martensitic transition (see for example Refs. 15–19). The process of nucleation and growth across a martensitic transition has been studied over many decades, and these MSM materials (besides their relevance to applications) provide potentially important systems for such studies with magnetic field H as an additional parameter. Recently, in a significant development it has been shown that the liquid to crystal transformation kinetics in monatomic Ge can be inhibited by the “magic ingredient” of pressure²⁰ and for magnetic systems it is magnetic field H which can be used as the “magic ingredient.” It is indeed found that H as a second control variable has a decisive role both for the magnetic first-order transition and on the kinetic arrest of such transformation process.²¹

The first-order transition as well as the associated supercooling and superheating can be depicted by lines in the H - T space for the magnetic materials. Similarly, the arrest of kinetics also can be represented by a line for a specific cooling rate, which can allow construction of the H - T phase diagram

for first-order transformation combined with the process of its kinetic arrest. In real multielement materials the accompanying quenched disorder broadens the sharp first-order transition.²² Consequently, the transition line and supercooling as well as superheating lines broaden into bands in H - T space, consisting of a quasicontinuum of lines. Each line corresponds to a region of the sample with length scale of the order of correlation length. Following the same argument, the H - T dependent kinetic arrest is depicted as a band in the phase diagram and is justified from the phenomenological studies.^{8,10,23} The broadening helps in using the second parameter H to produce the quasicontinuum of states of coexisting phases having different fractions of equilibrium phase and kinetically arrested high-temperature phase at low temperature, which vary with time, indicating metastability.^{4,9-11,24}

Thus, according to the phenomenological phase diagram, it becomes possible by traversing different H - T paths to initiate nucleation of the martensitic phase at different temperatures, much below its martensitic transition temperature, and study the growth of the equilibrium phase (martensitic). Here we have used a MSM alloy of composition $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$ and we study the growth of equilibrium phase (martensitic) with time at 25 K and 50 K starting with different phase fractions of nonequilibrium austenite phase created by traversing different H - T paths. We show that, in the standard process of cooling and measuring in 4 T field, the rate of growth of the martensitic phase is drastically less at 25 K compared to the 50 K in spite of having a similar starting fraction of the nonequilibrium austenite phase, indicating the effect of kinetic arrest of the first-order transformation.^{4,15} On the contrary, the growth rate is found to be similar at 25 K and 50 K (but significantly higher compared to the previous case) when the sample was cooled in 8 T to the respective temperatures and the measurement field is isothermally reduced to 4 T. Further, it is found that although the growth rate depends on the initial nonequilibrium phase fraction, it has very weak dependence on temperature for this

protocol. It is shown that while cooling in 8 T, nucleation of the martensitic phase is completely inhibited and the system approaches 50 K or 25 K with fully arrested austenite phase. The nucleation of the martensitic phase starts only while reducing the field, whereas, while cooling in 4 T, the nucleation of the martensitic phase has started at much higher temperature (≈ 150 K) and the system reached 25 K or 50 K with substantial fraction of transformed martensitic phase. An attempt is made to explain the intriguing time evolution through the motion of the interface between the austenite and martensitic phases.

II. EXPERIMENTAL DETAILS

The ribbon sample of MSM alloy of composition $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$ (Sn-12) was prepared by melt-spinning of the pre-melted alloy prepared from high-purity elements under a high-purity argon atmosphere. The composition and the crystal structure were determined from energy-dispersive x-ray spectroscopy (EDXS) and powder x-ray diffraction (XRD) techniques, respectively. The details of the sample preparation and characterization are given in Ref. 19. The

magnetization measurement was carried out in a commercial 14 T VSM (PPMS) made by Quantum Design, USA. For magnetic measurements, cooling or heating was always done at the rate of 1.5 K/min and the field changing was done at the rate of 100 Oe/sec. The time decay measurements were done immediately after temperature stabilization or field change. The M - H measurements are performed while sweeping the field at the same rate.

III. RESULTS AND DISCUSSION

Magnetization M as a function of T for $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$ is shown in Fig. 1(a). After cooling the sample in zero field from 350 K, a 4 T field was switched on at 5 K and M is measured while warming for the zero-field-cooled (ZFC) branch. Then M is measured while cooling in the same field for field-cooled cooling (FCC) and while warming after field cooling (FCW). The FCC and FCW branches show a paramagnetic to ferromagnetic (austenite) transition at high T followed by a hysteretic austenite to martensite first-order transition at lower T . Although the thermal hysteresis in $M(T)$

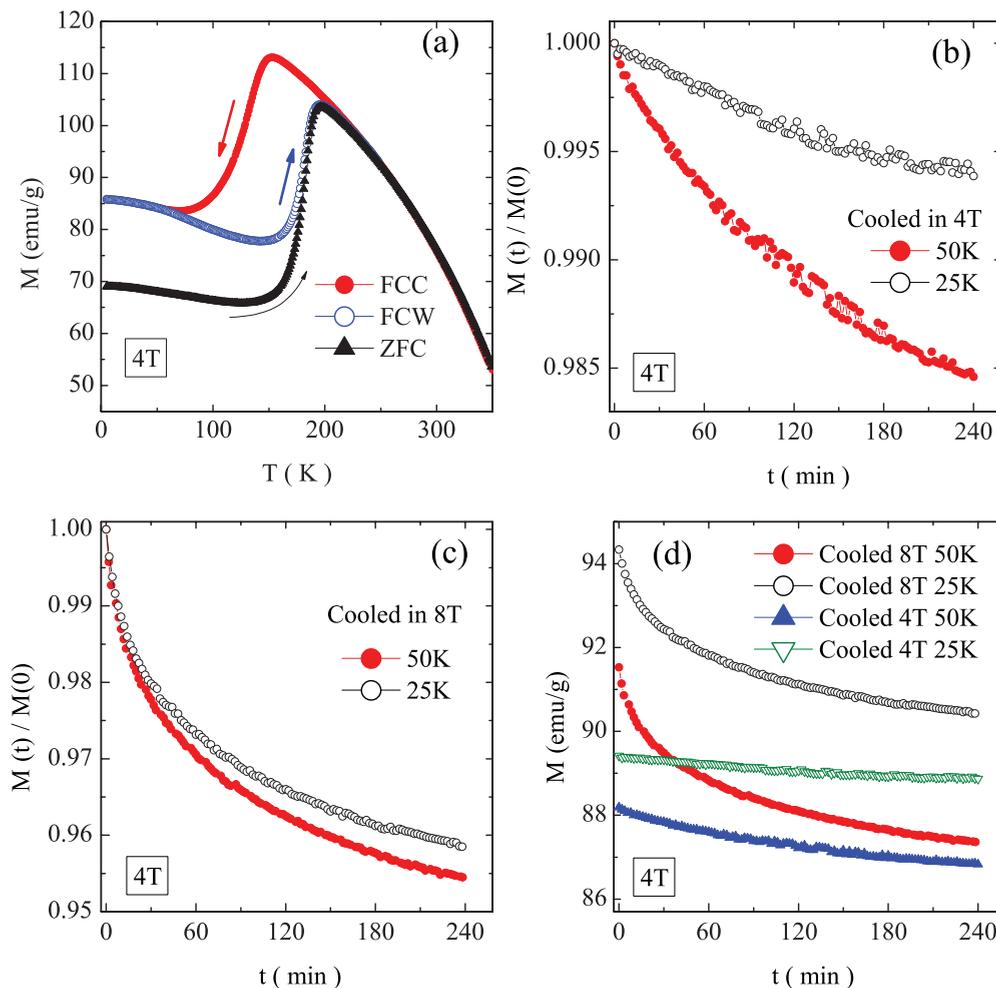


FIG. 1. (Color online) Magnetization M as a function of temperature T and time t measured at 4 T field following various protocols for $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$. (a) M vs T for ZFC, FCC, and FCW paths. (b) Normalized M vs t at 25 K and 50 K after cooling from 350 K in 4 T. (c) Normalized M vs t at 25 K and 50 K after cooling from 350 K in 8 T and isothermally reducing the field to measuring field of 4 T at the respective temperatures. (d) Decay of absolute value of M as a function of t .

closes around 65 K, the bifurcation between ZFC and FCC (or FCW) branches indicates that the first-order transition is not completed down to the lowest T . This is a clear indication of the presence of kinetically arrested high- T austenite phase, at low T .^{12,15,16} In a recent study on a Ni-Mn-In-Co system it is shown that there is metastability while cooling across the first-order austenite to martensitic transformation whereas there is no metastability while heating.²⁵ This indicates that there is supercooling but no superheating in this system. This asymmetry between superheating and supercooling is commonly observed in the case of melting of solids where nucleation occurs on the surface.²

To probe the low- T state, we measured M as a function of time t , $M(t)$, at 25 K and 50 K by reaching the respective measurement temperatures by two paths in H - T space: one by cooling from 350 K in 4 T down to 25 K or 50 K and measured $M(t)$. Figure 1(b) shows the $M(t)$ vs t after normalization with the respective M (at $t = 0$), $M(0)$, for the respective measurement temperatures. In the second protocol, the same points in H - T space are reached by cooling from 350 K in 8 T down to 25 K or 50 K and then isothermally reducing to the measurement field of 4 T. Figure 1(c) shows the normalized $M(t)$ vs t for 25 K and 50 K. It may be noted that although both the measurement temperatures are below the closure of thermal hysteresis related to first-order austenite to martensitic transformation, the magnetic states at 25 K or 50 K are far from equilibrium. The decrease in M with t indicates that the low- M martensitic phase grows from the high- M metastable austenite phase fraction. However, this growth of martensitic phase has the following rather intriguing features:

(1) The rate of growth is higher for 50 K compared to 25 K for both protocols. In the first protocol of measurement, i.e., cooling and measuring in 4 T, the decay in M at 50 K is almost 3 times that of 25 K. This is contrary to the transformation process related to first-order transition, in which case the barrier in free energy decreases as the system approaches the supercooling limit leading to faster growth of the low- T equilibrium phase with the decrease in temperature. On the contrary, the observed trend in the growth rate is similar to the kinetically arrested state indicating that the austenite phase fraction is in a nonequilibrium glasslike state at these temperatures.^{4,12,24}

(2) The decay in M is not directly related to the initial magnetization value as shown in Fig. 1(d). Although the 4-T-cooled 25 K measured state has intermediate starting value of M it shows the slowest decay.

(3) The most intriguing aspect is the significantly different rates of growth of the equilibrium martensitic phase at the same temperature and measuring fields, i.e., 4 T and 25 K or 50 K. While in the first protocol [Fig. 1(b)], the magnetization decreases by 0.6% over 4 hours at 25 K, the same is about 1.6% for 50 K. In contrast to that the magnetization decreases by more than 4% over 4 hours for both 25 K and 50 K in the second protocol [Fig. 1(c)].

It was theoretically predicted and also experimentally verified for the vortex matter that the region of metastability depends on traversed paths in two-variable space.^{26,27} Although this may offer some justification to the observed drastically different relaxation rates for the two paths (point 3), the slower relaxation at lower temperature (point 1) cannot be explained

only on the basis of the first-order transformation process. Since, for a first-order transition, the free energy barrier height decreases with the decrease in temperature, as the system approaches the supercooling limit, the relaxation rate increases with decrease in temperature. However, the observed behavior is a typical signature of the glasslike arrested state. For a glassy system the relaxation rate becomes critically slow with the decrease in temperature as is also observed for the “magnetic glass.”^{4,15,24}

To comprehend these observations we measured $M(t)$ at 25 K and 50 K in different H after cooling from 350 K in 8 T and isothermally reducing the field to the measurement field at the respective measurement temperatures. $M(t)$ for 25 K and 50 K are shown in Figs. 2(a) and 2(b), respectively. While the growth rate of the martensitic phase for all the measurements $H \leq 7$ T are high (more than 4% decay of M over 4 hours) at 50 K [Fig. 2(b)], a similar growth rate is observed only for the measuring $H \leq 5$ T at 25 K [Fig. 2(a)]. The growth rate of the martensitic phase at 25 K is rather low for 7 T; less than 1% decay in M is observed over 4 hours. The same is higher but less than 3% for 6 T measuring H . This apparently puzzling behavior appears to be related to the H at which the nucleation of the martensitic phase begins while reducing the field from 8 T at the respective temperatures. Figure 2(c) shows the isothermal field reduction M - H curves at 15 K, 25 K, and 50 K after cooling the sample in 8 T. The sharp fall in M while reducing the H from 8 T marks the onset of the nucleation process of the martensitic phase. At 50 K the nucleation starts around 7.5 T and for 25 K it is around 6 T. This can be justified from the schematic H - T phase diagram shown in Fig. 2(d), which is similar to the phenomenological phase diagram proposed initially for the high- T ferromagnetic phase [see Fig. 1(a) of Ref. 8]. While cooling in 8 T following path 2, a large fraction of the supercooled austenite phase gets arrested because various regions encounter the kinetic arrest (T_K, H_K) band before crossing the corresponding supercooling limits (T^*, H^*). When the field is reduced at 50 K or 25 K, the system traverses the (T_K, H_K) band from the opposite side and the arrested phase fractions gets progressively de-arrested resulting in the nucleation of the martensitic phase. Since at 50 K, the (T_K, H_K) band is encountered at a higher field than the 25 K path, nucleation of the martensitic phase starts at a higher field for 50 K. This explains the observed time evolution of M indicating the growth of martensitic phase at 25 K and 50 K for different fields [Figs. 2(a) and 2(b)]. The decrease in M is faster when the measuring field is lower than the field for onset of nucleation; above that field there is hardly any nucleation or time evolution of M and around this field there is intermediate rate of increase in the nucleation of the martensitic phase.

We attempt to unify the observed H - T path dependent diversity by quantitative analysis of the time dependent evolution of the equilibrium phase (martensite) from the nonequilibrium phase (austenite) at 25 K and 50 K for path 2. Let $P_{Eq}(t)$ and $P_{Neq}(t)$ be the equilibrium and nonequilibrium phase fractions at time t and saturation magnetizations of the two states are denoted as M_{Eq} and M_{Neq} , respectively. If $P_{Eq}(0)$ is the starting equilibrium phase fraction then $P_{Neq}(0) = [1 - P_{Eq}(0)]$ is the starting nonequilibrium phase fraction.

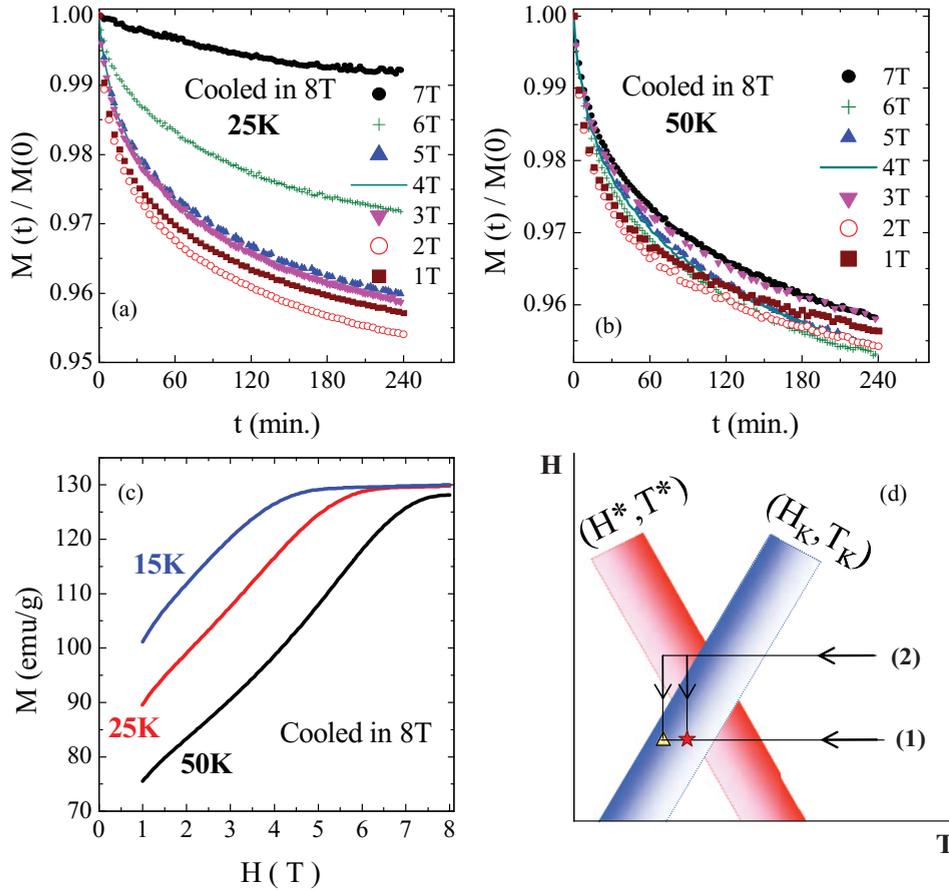


FIG. 2. (Color online) Time and H dependence of M after cooling in 8 T from 350 K and schematic H - T diagram for $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$. (a) Normalized M vs t at 25 K after cooling from 350 K in 8 T and isothermally reducing to different measuring H . (b) M vs t at 50 K following the same protocol as (a). (c) M vs H at 15 K, 25 K, and 50 K while reducing the field after cooling from 350 K to respective measurement T in 8 T. (d) Schematic H - T diagram depicting supercooling (H^* , T^*) and kinetic arrest (H_K , T_K) bands and also the two prominent paths followed in the present study.

Since broad distribution of relaxation rates leads to a logarithmic relaxation,²⁸ we consider a logarithmic time decay of the nonequilibrium austenite phase as

$$P_{Neq}(t) = P_{Neq}(0) [1 - D \ln(t/t_0)], \quad (1)$$

where D is the rate constant. It can be shown that the magnetization at any time t is given by

$$M(t) = M(0) - [1 - P_{Eq}(0)] \Delta M D \ln(t/t_0), \quad (2)$$

where $\Delta M = M_{Neq} - M_{Eq}$, and the starting magnetization value $M(0)$, which can be shown to be

$$\begin{aligned} M(0) &= P_{Eq}(0)M_{Eq} + [1 - P_{Eq}(0)]M_{Neq} \\ &= M_{Neq} - P_{Eq}(0)\Delta M. \end{aligned}$$

The zero-field-cooled state is the fully converted martensitic phase whose M (vs H) attains technical saturation around 1 T and does not undergo reverse martensitic transformation even up to 14 T field.¹⁹ Hence, the magnetization value at the respective fields (for $H > 1$ T) from the zero-field-cooled M - H curves (not shown here) at the measurement temperatures (25 K and 50 K) can be taken as corresponding M_{Eq} . When the system is cooled in 8 T it reaches 50 K or 25 K with fully arrested austenite phase, which is also shown to be a soft ferromagnetic phase having technical saturation around 1 T. Hence, extrapolation of the M - H curves of 25 K or 50 K [Fig. 2(c)] to the respective measurement fields (>1 T) gives the corresponding M_{Neq} values. These values along with the corresponding starting measured magnetization values, $M(0)$,

are used to obtain the two phase fractions and thus $P_{Eq}(0)$. The rate parameter D is the only fitting parameter for the fitting of Eq. (2) to the M vs t data at 25 K and 50 K [Figs. 2(a) and 2(b)]. These fittings are shown in Figs. 3(a) and 3(b) (fittings for only the alternate field values are shown for clarity), which indicate that Eq. (2) gives a reasonable description (for $t > 100$ sec) for the time evolution of the coexisting phases of this system. It may be noted that small deviation from the logarithmic behavior is experimentally observed in the relaxation of electrical resistance of a MSM alloy of the Ni-Mn-In-Co system.²⁵ Moreover, it is shown from rigorous theoretical calculations that logarithmic relaxation is valid only in certain asymptotic limits and for this case it is for the larger time (beyond 100 sec).^{28,29}

The above mentioned quantitative analysis brings out some intriguing aspects as shown in Fig. 3(c). Although the 3 T and 7 T curves show similar decay of total normalized magnetization at 50 K [as shown in Fig. 2(b)], they have very different fractions of nonequilibrium phase. After cooling in 8 T to 50 K when the field is reduced to 7 T, it has about 80% of the nonequilibrium phase whereas at 3 T the nonequilibrium phase fraction is only about 25%. Thus the normalized decay rate of the nonequilibrium phase fraction is significantly lower at 7 T compared to 3 T as shown in Fig. 3(c). This indicates a significant dependence of the decay rate D on the starting nonequilibrium phase fraction. Hence, we plot the decay rate D as a function of the starting nonequilibrium phase fraction $[1 - P_{Eq}(0)]$ in Fig. 4. It is rather significant that D appears to follows some kind of scaling with $[1 - P_{Eq}(0)]$ which may

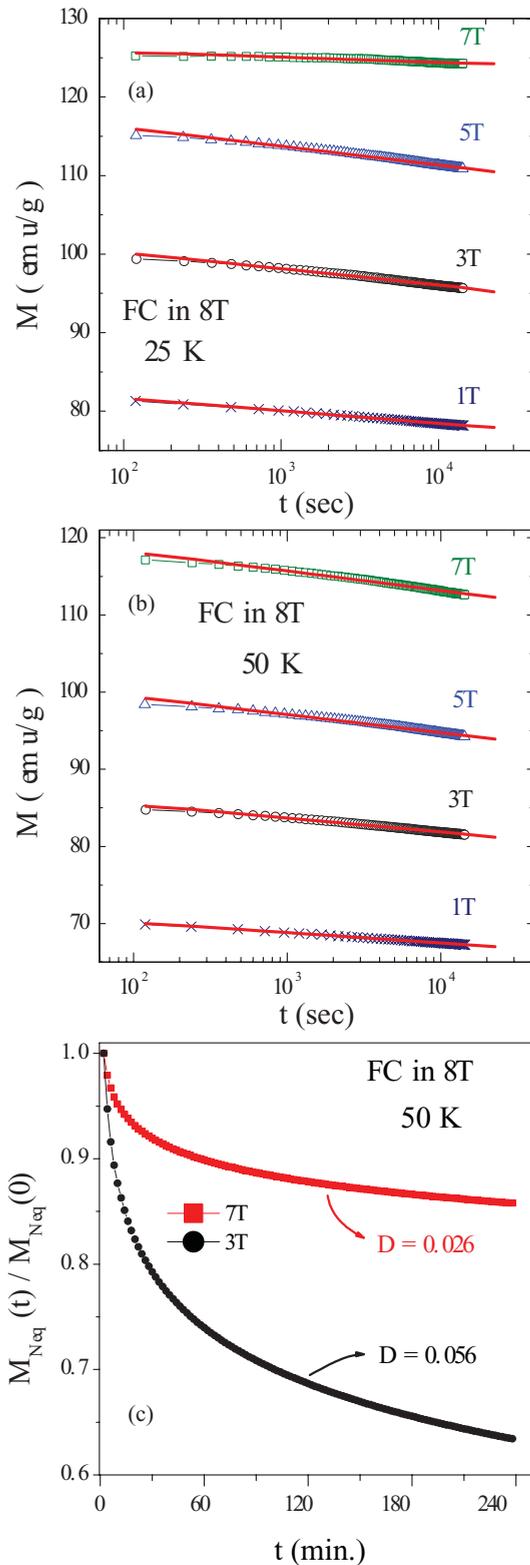


FIG. 3. (Color online) Time dependence of M at 25 K and 50 K in different measuring H after cooling in 8 T for $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$. (a) and (b) Logarithmic time dependence of M as well as the one-parameter fitting to Eq. (2) at 25 K and 50 K, respectively. Data and fitting for only the alternate H values are shown for clarity. (c) The calculated normalized nonequilibrium phase fraction vs t at 50 K for 3 T and 7 T after cooling in 8 T. The data are taken from Fig. 2(b) and the rate parameter D is taken from the fitting to Eq. (2).

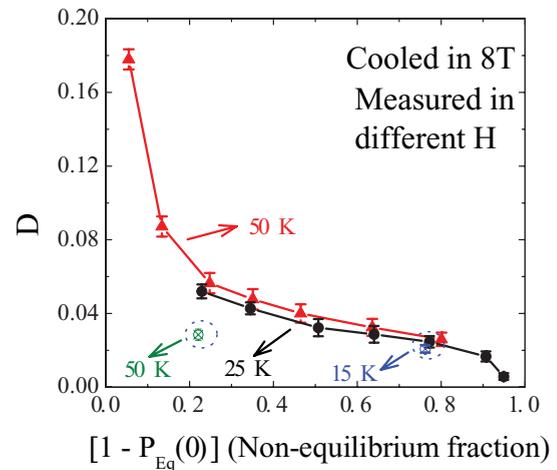


FIG. 4. (Color online) Variation of decay rate D of Eq. (2) as a function of starting nonequilibrium fraction $[1 - P_{Eq}(0)]$ for different paths in H - T space. The closed circles (black) and up triangles (red) are derived from the fittings of Figs. 3(a) and 3(b). The two points related to the fitting of M vs t by Eq. (2) for the following two other paths are also shown by open symbols with error bars encircled with dotted lines: (i) cooling in 4 T to 50 K (green); (ii) cooling in 8 T to 15 K, then isothermally reducing the field to 4 T (blue).

have interesting consequences for broad classes of materials. However, the decrease of D with increase in $[1 - P_{Eq}(0)]$, as well as its very weak temperature dependence, is a matter of real concern.

The nonequilibrium phase fraction is related to the “degree of metastability” and for a first-order transition this dictates the kinetics of the growth of the equilibrium phase.³⁰ In the initial stage of transition, the total volume of the nuclei of the equilibrium phase is rather small and their formation as well as growth is not correlated, since it is arising from the fluctuation in energy; hence, the effect on the “degree of metastability” is rather limited. With larger volume fraction of the equilibrium phase, the nature of the growth process, which is now correlated, is very different. However, in the present case the nucleation and growth processes of the equilibrium phase encounter two opposing effects as depicted by the overlapping supercooling and kinetic arrest bands [Fig. 2(d)]. Notwithstanding this complication, it offers us significant control on the process of nucleation by the second control parameter H and allows us to initiate nucleation at much lower temperatures by traversing different H - T paths. Recently, it has been shown for a CMR manganite that even for the same degree of metastability or the same fraction of nonequilibrium phase the rate of growth depend on the H - T history.³¹ This is attributed to the H - T path dependent critical radius of the nuclei leading to different sizes of mesoscopic domains whose distinct growth rate is dictated by the relaxation of the interfaces. The importance of the motion of the structural interface between two magnetic phases for the first-order transition and its kinetic arrest leading to a glasslike state at low temperature is highlighted through an experimental study on another CMR manganite system.³² Further, this study relates the motion of the interface of the structurally dissimilar phases with the growth of martensitic phase of shape memory alloys.

In the present case, while cooling in 8 T the nucleation is prevented by the process of kinetic arrest and it is initiated only at a much lower temperature by reducing the field. For example, while cooling in 4 T, nucleation of the martensitic phase starts at ≈ 150 K, whereas after 8 T cooling it is started only at the measurement temperature of 50 K or below. It is known that the critical radius R_C for nuclei formation is dictated by the difference between the equilibrium transformation temperature (≈ 200 K) and the nucleation temperature.^{30,33,34} After cooling in 8 T to the respective measurement temperature when field is reduced isothermally, then the de-arrest of the arrested austenite phase start below a certain H with the formation of the nuclei of the martensitic phase of critical radius R_C which populate as the field is progressively decreased. Thus, the starting nonequilibrium phase fraction is inversely related to the number of martensitic nuclei when the system is subjected to the time relaxation keeping the field constant. We expect that the growth process is dictated by the interface area of the initial state.³² This explains the observed scaling of the relaxation rate D with the nonequilibrium phase fraction of Fig. 4, for path 2. Observation of almost the same relaxation rate D for the same nonequilibrium phase fraction for 25 K and 50 K indicates that the R_C is similar for both the temperatures and the D is governed only by the number of martensitic nuclei or the starting nonequilibrium phase fraction. It is reasonable to expect that since the nucleation is prevented at the normal first-order transformation process, the critical radius has reached the lowest physical dimension and loses further temperature dependence. This conjecture is tested by measuring the time relaxation after cooling in 8 T to 15 K and isothermally reducing the field to 4 T. It is rather significant that even at 15 K the relaxation rate scales with the starting nonequilibrium phase fraction [calculated from 15 K M - H of Fig. 2(c)] and merges with curves of 25 K or 50 K within the error bars as shown in Fig. 4. Thus, for path 2, it can be safely asserted that relaxation at or below 50 K will follow the same scaling with the starting nonequilibrium phase fraction and fall on the same curve of Fig. 4.

The above-mentioned analysis is reconfirmed by calculating the relaxation rate for the 4-T-cooled state at 50 K. For this case, a larger fraction of the austenite phase has already converted to the equilibrium martensitic state while cooling in 4 T and the nucleation has started at ≈ 150 K [as shown in Fig. 1(a)] having larger critical radius R_C . Thus this state has larger size domains of the martensitic phase compared to the state created by cooling in 8 T and reducing the field at 50 K. Hence, for this 4-T-cooled state, the starting interface area is smaller for the same fraction of the nonequilibrium phase compared to the state created by path 2, resulting in a significantly smaller decay rate as show in Fig. 4. The D value for this 4-T-cooled state is much below the scaling curve of 50 K or 25 K (as shown in Fig. 4) and reinforces the

above-mentioned analysis. Moreover, this explains the reason behind the observed drastically different growth rates for path 1 and path 2 (Fig. 1). Hence, intense theoretical efforts backed by experiments on a variety of such magnetic systems where arrested kinetics give rise to glasslike long-range-ordered magnetic states are essential for deeper understanding of the apparently intriguing observations presented here. Further, such path dependent metastability is expected to be rather ubiquitous and needs to be investigated in other materials identified as “magnetic glasses” (see for example Ref. 35).

IV. CONCLUSION

We show that low- T metastable magnetic states in a shape memory alloy system, $\text{Ni}_{45}\text{Co}_5\text{Mn}_{38}\text{Sn}_{12}$, decay with very different rates at the same measuring field and temperature depending on how that H and T are reached from the high-temperature side. In this system the H - T induced broad first-order magnetic transition is inhibited by arrest of transformation kinetics resulting in persistence of the high- T austenite phase fraction which coexists at low T with the equilibrium martensite phase. The fractions of these phases depend on the H - T paths; however, magnetization of the metastable coexisting phase shows glasslike decay at low T . This study reveals that the decay rate of the metastable coexisting phase scales with the starting nonequilibrium phase fraction but is almost independent of temperature when the decay is studied after isothermal field reduction at a temperature much below the closure of the hysteresis related to the first-order transition or much below the martensitic transformation process, whereas the decay rate is significantly small at the same H and T when the standard process of cooling and measuring in the same field is followed. This apparently anomalous behavior is explained on the basis that the nucleation starting much below the first-order transition temperature has the smallest possible critical radius R_C of the nuclei of the martensitic phase and consequently larger interface area for the same fractions of phases compared to the case when the nucleation is started at a much higher temperature having much larger R_C . The long-time relaxation at low temperature takes place with the growth of the respective nuclei (or domain of the martensitic phase) and is related to the available starting interface area. Further, this study indicates that when the process of nucleation is inhibited by the arrest of kinetics allowing nucleation at much lower temperature by reduction of H , the size of the critical radius is independent of the temperature.

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