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<td>Lakhani, Archana; Dash, S.; Banerjee, A.; Chaddah, P.; Chen, X.; Ramanujan, R. V.</td>
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Tuning the austenite and martensite phase fraction in ferromagnetic shape memory alloy ribbons of Ni45Co5Mn38Sn12

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Tuning the austenite and martensite phase fraction in ferromagnetic shape memory alloy ribbons of Ni$_{45}$Co$_{5}$Mn$_{38}$Sn$_{12}$

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The ferromagnetic shape memory alloy ribbons with composition Ni$_{45}$Co$_{5}$Mn$_{38}$Sn$_{12}$ are shown to have field induced kinetically arrested ferromagnetic austenite phase down to the low temperature due to hindered martensite transformation. This gives rise to the coexisting martensite and austenite phases in a wide range of temperature and field. Here, we show a systematic rise in arrested austenite phase with the reduction in martensite phase quantitatively by various magnetization measurements. The fraction of these coexisting phases can be tuned in “field-temperature” space. Further, we show that the “domain” of tunability varies with temperature. © 2011 American Institute of Physics.

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Magnetic field induced phenomena like colossal magnetoresistance, magnetocaloric, and magnetic shape memory effects have motivated the development of new technology. Ferromagnetic shape memory alloys (FSMAs) with general formula Ni$_{50}$Mn$_{50}$X$_{y}$ (X = In, Sn, and Sb) belong to the class of magnetic materials having interesting functional behavior associated with the application of magnetic field.1–4 The functional performance of FSMAs as transducers, actuators, and switching devices is associated with the first order structural transition known as martensitic transition (MT). The high temperature phase is a ferromagnetic austenite (FM-A) phase, while the low temperature phase is low magnetization martensite (LM-M) phase, which could be either ferromagnetic or antiferromagnetic in nature. This magnetic-structural transition is very sensitive to the field, pressure, and composition, which makes them a good candidate for multifunctional applications.5

In these alloys, the first order MT is influenced by disorder which gives rise to the coexisting austenite and martensite metastable states. Metastability associated with phase co-existence in a system is an interesting feature which introduces the possibility of tuning a particular phase fraction with the application of external parameters like temperature, field, and pressure. This quantitative manipulation is important in the development of first order phase transition materials towards the aim of practical applications like in magnetic refrigeration and switching devices,6 because the path followed in the field-temperature (HT) space is a crucial criterion in deciding the final state of the system.

In this letter, we emphasize the tuning of phase fraction in Ni$_{45}$Co$_{5}$Mn$_{38}$Sn$_{12}$ alloy ribbons by magnetization measurements. Use of magnetocaloric materials in the form of ribbons enhances the technical performance of refrigerators.4 The sample used in the present investigation is the same one used for our earlier study;7 therefore, preparation and characterization can be seen from this reference. Magnetization measurements are carried out by using 14 T Physical Property Measurement System (PPMS)-Vibrating Sample Magnetometer (VSM) from Quantum Design, USA.

Figure 1(a) shows magnetization as a function of temperature while field cooled cooling (FCC) and field cooled warming (FCW) in various constant fields ranging from 0.05 T to 9 T marked on the respective curves. This data were reported in our earlier paper7 and is reproduced here for completeness. At 0.05 T, on decreasing the temperature, we observe a broad first order transition from FM-A to LM-M at martensitic transformation temperature (T$_{m}$), while on heating a reverse MT to the FM-A phase takes place. The characteristic temperatures for the onset and finish of martensite (M$_{s}$ and M$_{f}$) and austenite transition (A$_{s}$ and A$_{f}$) are marked on the 0.05 T curve of Figure 1(a). For 0.05 T, the phase co-existence region lies between ~210 K to ~70 K during cooling and heating cycles. This broadening of first order transition is due to the disorder induced by chemical inhomogeneities.8,9 On increasing the field, the hysteresis suppresses and T$_{m}$ decreases gradually. Here, T$_{m}$ is defined as T$_{m} = [(M_{s} + M_{f})/2 + (A_{s} + A_{f})/2]/2$. Variation of T$_{m}$ with increasing field is shown in the inset of Figure 1(a). The observed average shift in T$_{m}$ is ~4 K/T which demonstrates the high sensitivity of this alloy with field. This definition of T$_{m}$ is different from the T$_{c}$ defined as the temperature of the peak magnetization in our earlier study,7 and this is the cause of the lower value of dT$_{m}$/dH quoted here in comparison to dT$_{c}$/dH.

At low field ~0.05 T, the transition is broad and the magnetization at 5 K and 350 K is nearly same indicating almost a complete transformation from austenite to martensite-phase while cooling. At higher fields, this difference of magnetization at 350 K and 5 K increases, which is seen by a gradual rise in the value of magnetization at 5 K, indicating the gradual inhibition of martensitic transformation, and at 9 T, the MT is almost hindered. Hence, we see that the high temperature FM-A fraction remains frozen on increasing the magnetic field and we get coexisting phases of...
Cobalt derivatives 10–16 which confirm the metastable 41 emu/gm, while at 9 T, it is/C24 curve at 5 K,7 shown in the inset of Figure 1(b), we can estimate the arrested LM-M state as seen by the zero field cooled (ZFC) M(H) field. Since on cooling in zero field down to 5 K yields a reduction in the transformed martensite fraction with rise in field. The amount of ferromagnetic fraction depends on the martensite fraction across the transition decreases and arrested fraction increases giving rise to co-existing FM-A and LM-M states. The two curves are in very good one to one correspondence with each other as the sum of the transformed and arrested fraction at each field is ~100%. At 4 T, austenite and martensite fraction is almost equal across the transition.

To elucidate the arrested kinetics in magnetic systems, an unusual and now established measurement protocol in which cooling and heating in unequal fields (CHUFs) is performed. This protocol facilitates identifying glass like arrested state in various functional magnetic systems known as “magnetic glasses” by heating in different fields than the cooling fields by showing the evidence of de-arrest.7,17 Normally, there are two threshold field values H1 and H2 which decide the state of material at low temperature while cooling above and below these fields.18,19 In this case, since the low temperature state is a LM-M state, cooling above H2 (~8 T) results in the maximum arrested phase, while cooling below H1 (~1 T) results in the maximum transformed phase. The best field values for CHUF measurements are between H1 and H2, which give varying coexisting phase fractions when cooled in various fields above and below these values. We have performed various magnetization measurements in a range of measured fields after cooling in various fields; here as an example, we show M(T) measurements at 4 T, in Figure 2, after cooling in various fields Hcool = 0, 1, 2, 3, 4, 5, 6, 7, and 8 T. In each case, the sample is cooled from 350 K down to 5 K in field, and then field is changed isothermally to 4 T at 5 K. At 4 T, austenite and martensite phase fraction is 50% each, as seen from Figure 1(b). When Hcool ≤ 4 T, only one transition is seen, this corresponds to reverse MT at ~200 K. On the other hand, for Hcool > 4 T, there are two transitions, one indicated by sharp fall in magnetization while heating and the other by rise in magnetization at ~200 K. The sharp fall in magnetization after cooling in lower fields signifies the de-arrest of arrested FM-A fraction, while the rise in magnetization indicates the reverse martensite transformation to FM-A state.

The magnetization value at 5 K increases gradually on increasing the cooling fields, indicating the larger arrested fraction at low temperature on cooling in higher fields. Taking the sum of arrested and transformed fraction at 9 T as

![FIG. 1. (Color online) (a) Temperature dependent magnetization for Ni45Co5Mn38Sn12 at various fields as shown on the respective curves (reproduced from Ref. 7). Inset shows the variation of Tm with field. (b) Arrested (FM-A) and transformed (LM-M) phase fraction as a function of field at 5 K. Inset (taken from Ref. 7): Field dependent magnetization at 5 K after cooling in zero field.](image1)

![FIG. 2. (Color online) Magnetization w.r.t T, while warming in 4 T after cooling in different fields from 0–8 T. Data are reproduced from Ref. 7. Inset shows the arrested phase fraction as a function of cooling field at 4 T.](image2)
100% arrested FM-A fraction from FC-M(T) measurements as above, we have calculated the corresponding arrested phase fraction as a function of cooling field from CHUF magnetization data at 4 T, which is shown in the inset of Figure 2. The arrested fraction increases up to 90% at 4 T on increasing the cooling field to 8 T. It shows that the field induced state has the memory of last field experienced. This is a paradigm of magnetic memory effect seen in these alloys where a variety of coexisting phase combinations can be shaped by varying the values of cooling fields. Similarly, at various measuring fields, the arrested fraction increases on increasing the cooling fields as shown by the vertical lines in Figure 3(a). The rectangular area represents the domain of arrested phase fraction at 5 K. In the same way, we have calculated the arrested phase fraction as a function of measuring field after cooling in various fields at 25 K, 50 K, and 75 K shown in Figures 3(b)–3(d), respectively. Evidently, the area representing the arrested fraction decreases with increase in temperature from 5 to 75 K indicating that the “domain” of tunability of phase fraction in a range of field depends on temperature. Hence, for FSMAs, we can tune the austenite and martensite phases in the HT space. This feature can be utilized for the device applications.

Since phase co-existence is the key parameter for associated functionalities in metamagnetic materials, the understanding of path dependence in the phase space defined in terms of controlled variables like magnetic field and temperature can be helpful in understanding the use of these sensitive functional alloys as potential magnetic refrigerants and switching devices.6,20

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