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Review Paper

A Short Review of Magnetocaloric Effect in Ni-Mn-Ga Heusler Alloy System

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Abstract— Magnetic refrigeration that utilizes the magnetocaloric effect (MCE) of a material is considered a promising substitute to the conventional gas-compression/expansion cooling technology owing to its advantages, such as environmental friendliness, cost-effectiveness, etc. For the potential application of this technology, low-cost and highly efficient magnetocaloric materials are in great need as magnetic refrigerants. The geometry of the magnetocaloric materials also becomes important for cooling in (nano)macro devices and it demands a very small size. In search of prospective magnetocaloric material Ni-Mn-based ferromagnetic Heulser alloys are of great interest for their potential to achieve large/giant magnetic entropy change at magneto-structural transition. This review article comprises an overview of the magnetocaloric effect in the ferromagnetic Ni-Mn-Ga Heusler alloy system. MCE in these alloys in various low/reduced dimensions such as ribbons, microwires and thin films are also outlined. Recent development in this field along with previous works have been reviewed in a systematic manner. The present difficulties/limitations and remaining challenges in this field have also been discussed in this article.

Keywords— Heusler alloy, Martensitic transition, Magnetic entropy, Magnetocaloric effect, Microwires, Ribbons, Thin films.

1. Introduction

During the last few years, Ni-Mn-based ferromagnetic Heusler alloys have attracted much attention due to their multifunctional properties. They exhibit large magnetic field-induced strain, large/giant magnetocaloric effect, magnetoresistance (MR) etc. In this respect many such materials have been developed recently, such as Ni-Mn-Ga [1], Ni-Fe-Ga [2], Ni-Mn-Al [3], Ni-Co-Al [4] etc. Off-stoichiometric composition of Ni-Mn-X (Z= In, Sn, Sb) Heusler alloys are the most significant magnetocaloric materials as they can produce large magnetic entropy change due to magnetic and magneto-structural transitions [5, 6]. The metamagnetic structural transition from the weak-magnetic martensite to ferromagnetic austenite under an applied magnetic field leads to an inverse MCE in these alloys [7]. Ni-Mn-Ga Heusler alloys have been studied extensively due to their large field-induced strain [8, 9], giant magnetocaloric effect [10, 11] and large magnetoresistance [12-14]. The alloys show a large/giant MCE associated with a strong coupling between magnetic and structural degrees of freedom near the magnetostructural transition. This magnetostructural transition is accompanied by changes in the crystal structure. Moreover, a tuneable MCE with a wide working temperature interval (WTI) can be achieved in this group of alloys by composition variation which makes them practically important [15, 16]. Recently, doping in Ni-Mn-based Heusler alloy by suitable materials rejuvenates the area of research in MCE [17,18]. Doping of non-magnetic elements such as Cu, Al, Ti, Cr, C, etc. and ferromagnetic elements like Co and Fe in these alloys also have been studied to enhance the MCE [19-22]. Besides large MCE some of these alloys exhibit large MR near the structural transition temperature on the application of high magnetic field [5, 12]. Different forms of the alloys like thin films, ribbons and microwires have been prepared to enhance physical or magnetic properties with respect to their bulk counterparts [14, 15, 23]. In this article, a detailed review of MCE in the Ni-Mn-Ga ferromagnetic Heusler alloy family with various compositions and different forms (ribbons, microwires and thin films) has been covered. The effect of doping with different elements and hydrostatic pressure on magnetocaloric properties has also been reviewed. Recent development in this field along with previous works have been reviewed in a systematic manner which may help the beginner in this research field.

The above discussions represent a brief description of Ni-Mn-based Heusler alloys. The rest of the paper is organized as follows; Section 2 contains the comparison of the magnetocaloric effect in various compositions of Ni-Mn-Ga

alloys. In section 3, comparative findings of the MCE in different forms of the alloys like ribbons, microwires and thin films have been depicted. The effect of doping and hydrostatic pressure on MCE in these alloys is also presented in this section. Section 4 encompasses the conclusion of this article and the future scope in this field.

2. Comparison

Magnetocaloric effect in various compositions of Ni-Mn-Ga alloys

During heating both stoichiometric and off-stoichiometric compounds with a composition near to Ni_2MnGa undergo a first-order structural transition from tetragonal martensite to cubic austenite and the reverse process on cooling [24, 25]. Almost all the functional properties such as magnetic field-induced strain, magnetocaloric effect, magnetoresistance etc. of these alloys are linked with the first-order magneto-structural transition (FOMST). These alloys have a cubic $L2_1$ structure in the high-temperature austenitic state and go through a sequence of intermediate various modulated-martensite structures on cooling. The austenite phase is a crystallographically stable cubic $L2_1$ structure (space group Fm3m) which consists of four interpenetrating fcc sublattices A, B, C and D with origins at (0, 0, 0), (1/4,1/4,1/4), (1/2,1/2,1/2) and (3/4,3/4,3/4). In which A and C sublattices are occupied by Ni atoms. B and D are occupied by Mn and Ga atoms respectively [26]. The probable martensite structures can be non-modulated (termed as NM) as well as modulated such as five-layer modulated (10 M), seven-layer modulated (14 M), six-layer modulated (12 M) martensite [27-29]. During cooling a first-order magnetic transition and its accompanying structural phase transition (Martensitic transition) occurs in this alloy. As the structural and magnetic phase transitions in these alloys are very sensitive to the composition, this opens up the opportunity for variations of the transition temperatures up to their coincidence. This careful coupling of those transitions is an improvement criterion for obtaining large MCE in Ni-Mn-Ga alloy systems [30]. With the implementation of this criterion, it has become possible to enhance MCE in this alloy by coupling structural and magnetic transitions [27]. During the last few years, large MCE in Ni-Mn-Ga alloy systems has been observed in single crystals [11], polycrystalline bulk alloys [30], melt-spun ribbons [15], microwires [31] and thin film [32] also. For the Ni–Mn–Ga alloy system, the transition from a low-temperature ferromagnetic martensitic phase to a high-temperature paramagnetic austenitic phase occurs, [12-14] whereas the Ni–Mn–X (X = In, Sn, and Sb) alloy system the transition arises from paramagnetic martensite to ferromagnetic austenite [33, 34]. Owing to this different characteristic of the magnetostructural transition, the Ni–Mn–X (X = In, Sn, Sb) alloys show an inverse magnetocaloric effect contrary to Ni-Mn-Ga alloys. At martensitic transition, the saturation magnetization between Martensite and austenite phase ($\Delta M=M_{Martensite}-M_{Austenite}$) is an important parameter responsible for large magnetic entropy change (ΔS_M) [5]. In a relatively small field, the martensite to austenite transition is attended by lower to higher magnetization but at a high field the opposite situation is observed. The variation of ΔM with an applied magnetic field for different Ni-Mn-Ga alloys has been reported in the literature [35, 36]. In this alloy system, ΔM is initially negative and shows a minimum at a particular field. For larger applied fields, ΔM increases with increasing field and reaches a positive saturation value ΔM_{sat} [37, 38]. Due to the large magnetocrystalline anisotropy constant (K_u) in the martensite phase a much higher applied magnetic field is required for saturation magnetization for this phase compared to the austenite phase. This magnetocrystalline anisotropy decreases with the increase of Ni content in off-stoichiometric Ni-Mn-Ga alloys as observed by Albertini et al [38]. In of-stoichiometric Ni-Mn-Ga Heusler alloy, the value of the saturation magnetization difference between the phases (ΔM) can be enhanced by composition variation, heat treatment and by adding suitable elements [37, 38]. It is well documented that the valence electron concentration e/a is a crucial feature that influences the martensitic transition temperature and the magnetic properties of the alloys as well as ΔM. The anisotropy constant (K_u) decreases with the increase of e/a [38]. It has been established that the monotonic decrease of ΔM is a consequence of a low magnetic anisotropy in the martensitic phase [39]. The shift of structural transition temperature (T_M) in Ni_2MnGa alloy was observed when Mn and Ni atoms were partially substituted for Ga. Studies of $Ni_{2+x}Mn_{1-x}Ga$ and $Ni_2Mn_{1+x}Ga_{1-x}$ alloys showed an increase of T_M and a decrease of T_C (Ferro- Para Curie transition) with the deviation from stoichiometry [40-42]. The increase of T_M in these alloy systems to the increase in valance electron concentration e/a is attributed due to the Hume-Rothery mechanism [25, 43]. The decrease of T_C as observed in $Ni_{2+x}Mn_{1-x}Ga$ and $Ni_2Mn_{1+x}Ga_{1-x}$ alloys has a different origin. Since in Ni-Mn-Ga alloys, the magnetic moment of ~4 μ_B is located on Mn atoms, lowering T_C in the alloy can be explained due to the dilution of the magnetic subsystem. Since the coincidence of T_M and T_C is an enhancement criterion of MCE in this alloy system, several studies have been reported for $Ni_{2+x}Mn_{1-x}Ga$ ($0\leq x\leq 0.20$) [44-46]. It was observed that the martensitic and ferromagnetic transition temperatures merged for x=0.18 [47]. A non-monotonous behaviour of shift of T_M with Ni-excess was also observed in these alloys. Giant MCE with $\Delta S_M = -20.4$ and -9.2 J kg-1 K^{-1} in fields of 5 and 1.5 T respectively at 317 K reported by Zhou et al. for $Ni_{55.2}Mn_{18.6}Ga_{26.2}$ alloy where first-order magnetic transition and its attendant martensitic structural phase transition co-inside with each other [44]. Studies in $Ni_{2+x}Mn_{1-x}Ga$ alloys with excess Ni content have gained special attention as T_M and T_C in these alloys can be merged by varying x and this coupled magneto-structural transition gives rise to a large MCE [48]. A series of alloys with nominal composition $Ni_{2+x}Mn_{1-x}Ga$ (x=0.16, 0.18, 0.20, 0.22, 0.24, 0.26) was studied by Mandal et al. to establish the coincidence of these two transformation temperatures [49]. It was observed that the T_M increases and T_C decreases with the increase of Ni content in $Ni_{2+x}Mn_{1-x}Ga$ alloy and these two transitions almost coincide for the composition $Ni_{2.208}Mn_{0.737}Ga$. On further increase of Ni content martensitic transition below T_C was not observed as structural transition crossed the magnetic transition ($T_M>T_C$). To determine the isothermal change of magnetic entropy (ΔS_M)

Maxwell's thermodynamic equation was used and a large ΔS_M of -96 J/kg K at 290.75 K was reported for the alloy $Ni_{2.208}Mn_{0.737}Ga$ at a magnetic field of 5T. However, using Clausius–Clapeyron equation ($dH/dT=-\Delta S_M/\Delta M$), a much lower value of ΔS_M ~ - 21 J/kg K was reported for this alloy. Recently, the magnetocaloric effect in $Ni_{2+x}Mn_{1-x}Ga$ alloy with coupled magnetostructural phase transition was reported by Khovaylo et al. [40]. Magnetic entropy change, ΔS_M and adiabatic temperature change, ΔT_{ad} in the $Ni_{2+x}Mn_{1-x}Ga$ ($0.18 \leq x \leq 0.27$) alloy system reported by them. A large ΔS_M of -25 J/kg K was observed in $Ni_{2.18}Mn_{0.82}Ga$ and $Ni_{2.20}Mn_{0.80}Ga$ alloys. With further increase of Ni excess, the ΔS_M decreases as magnetization saturation of $Ni_{2+x}Mn_{1-x}Ga$ decreases with a deviation from stoichiometry due to the dilution of the magnetic subsystem. It was observed that the total entropy change at the martensitic transformation increases in the range of composition of $0 \leq x \leq 0.19$, reaches a maximum for $x = 0.19–0.20$ (coupled magnetostructural phase transition) and decreases for $x \geq 0.22$. With a higher Ni excess, the magnetic entropy change in the alloys was found to be decreased considerably. With the increasing Ni content, a decrease in saturation magnetization in the martensite phase lower the value of ΔS_M. Adiabatic temperature change ΔT_{ad} ~ 0.8 K (during heating) and 1.8K (during cooling) was reported for the sample $Ni_{2.18}Mn_{0.82}Ga$ which showed the highest ΔS_M. For the other samples, ΔT_{ad} was found significantly small.

3. Comparative finding

3.1 Effect of doping on magnetocaloric properties and phase transition in Ni-Mn-Ga alloys

Doping with the suitable element in this alloy system has gained considerable attention to the researcher as it can modify the structural transition temperature and enhance the magnetocaloric effect. The effect of doping by elements such as Fe, Co, Cu, Pt, In, Si, Cr etc. on MCE and the structural transition has been studied extensively [50-57]. Soto et al. reported that the T_M and T_I shifted to lower values when 'Fe' was substituted in a nearly stoichiometric Ni-Mn-Ga alloy, whereas the Tc shifted towards higher temperatures [50]. It was suggested that the phase stability is not only governed by valance electron concentration (e/a) but there are other parameters also. The substitution of 'Co' in Ni–Mn–Ga Heusler alloys around the Mn-rich composition $Ni_{50}Mn_{30}Ga_{20}$ affects the transition temperatures and alters the exchange interactions of martensite and austenite phases reported by Fabbrici et al [58]. A reverse martensitic transformation from paramagnetic martensite to ferromagnetic austenite phase was observed by substitution of 'Co' to Mn-rich Ni-Mn-Ga Heusler alloys which in turn gives rise to an inverse magnetocaloric effect. The saturation magnetization moment of the austenite phase is strongly enhanced by Co compared to the martensite phase produces a large ΔM at the transition which is responsible for a large inverse MCE. They also reported a high value of dT_M/dH (shift of transition temperature per magnetic field) and estimated isothermal magnetic entropy change from the Clausius–Clapeyron equation. The refrigeration capabilities, estimated by the dT_M/dH values, show promising values for application purposes in comparison to other NiMnX alloys. Increases in the martensitic transformation temperatures and decreases of T_C with the addition of Cu to stoichiometric Heusler type Ni_2MnGa were reported by Sarkar et al [54]. Fine-tuning of M_S with T_C was achieved by Cu substitution in $Ni_{50}Mn_{25-x}Cu_xGa_{25}$ ($0 \leq x \leq 7.0$) alloy. Maximum Refrigerant Capacity (RCP) of 327.0 J/Kg was reported at 6.5 at% of Cu in this alloy for which a virtual overlap between T_C and T_A occurred at a temperature of 303 K. Shift of T_M to high values in these alloys was explained on the basis of e/a concentration. The addition of Cu decreases T_C as it replaces Mn atoms which carry the magnetic moment in this type of Heusler alloy. A large isothermal magnetic entropy change of 7 J/kg K at 288 K for 5 T was observed in Pt-doped Ni_2MnGa alloy [53]. Doping with Pt in Ni_2MnGa results in an increase in saturation magnetic moment. The increase in the magnetic moment was explained due to an increase in the Mn and Pt local moments. The large magnetic entropy change at 288 K was observed for $Ni_{1.8}Pt_{0.2}MnGa$ due to first-order magneto-structural transition from martensite to austenite in a ferromagnetic state. Recently, a large ΔS_M of −36 J/kg K under 5 T magnetic field was reported for $Ni_{50}Mn_{19}Ga_{25}Cu_6$ alloy at 300K with low hysteresis [56]. The magnetostructural transition between paramagnetic austenite and ferromagnetic martensite takes place for this composition giving rise to a giant magnetocaloric effect suitable for room-temperature refrigeration. A small amount of Co doping also affects the structural property of the alloy such as microstructure, grain boundaries, modulation in the martensite phase etc. With the increase of Co-doping, the grain size reduced, and the crystal structure of the martensitic phase became 2 M martensite due to the progress of γ-phase precipitates reported by Namvari et al [59].

3.2 Effect of hydrostatic pressure on magnetocaloric properties and phase transition in Ni-Mn-Ga alloys

It is desirable to enhance the magnetocaloric property of the magnetocaloric materials by external parameters like pressure, electric field etc. Pressure is an external parameter that can affect the structural property as well as the structural entropy of the alloy system. A significant number of efforts have been made to study the effect of hydrostatic pressure on MCE, structural and magnetic phase transitions in Ni-Mn-based Heusler alloys [60-69]. However, most of the studies showed that the structural and magnetic transition temperatures have a strong dependence on the pressure but there is no significant change in magnetic entropy in these alloy systems, rather it decreases with the increase of hydrostatic pressure. Application of pressure (8 Kbar) in $Ni_{2+x}Mn_{1-x}Ga$ (x=0.16, 0.18, 0.20, 0.22, 0.24, 0.26) alloys shifted the martensitic transition (T_M) and Curie temperature (T_C) at higher values reported by Mandal et al [30]. Application of hydrostatic pressure in these alloys slightly decreases isothermal magnetic entropy change (ΔS_M) however it reduces the hysteresis in the M vs H curve. The value of dM_S/dp and and dT_C/dp was found to be 0.60 K/kbar and 0.46 K/kbar respectively for the sample x=0.16. Similar results were also observed by Albertini et al [70]. The effect of hydrostatic pressure on the magnetic and magnetocaloric properties of stoichiometric Ni_2MnGa and $Ni_{1.85}Mn_{1.15}Ga$

Heusler alloys have been investigated by Devarajan et al [64]. It was observed that the application of hydrostatic pressure increased the T_M for Ni_2MnGa alloy whereas it decreased the same for $Ni_{1.85}Mn_{1.15}Ga$. The metamagnetic-like transition was suppressed in both specimens with increasing pressure. For both the samples the ΔS_M was decreased while applying pressure. The ΔS_M was found to reduce from 19.2 J /kg K (P= 0) to 6.04 J/ kg K (P= 9.69 Kbar) around martensitic transition for Ni_2MnGa alloy. The effect of pressure on the magnetic and magnetocaloric properties of Pt-doped Ni-Mn-Ga alloys was studied by Sivaprakash et al [68]. They reported an increase of T_M with the application of hydrostatic pressure (3.5K/GPa) for $Ni_{2-x}Pt_xMnGa$ (x= 0.2, 0.3 and 1.0) alloys. The hydrostatic pressure stabilized the martensite phase. The maximum magnetic entropy change (ΔS_{max}) was found to reduce from 9.31 J /Kg K to 5.52 J /Kg K at a hydrostatic pressure of 0.91 GPa while other parameters were unchanged. Pressure also induced the AFM/FM interaction at low temperatures for x = 0.2 alloy. The decrease of ΔS_M with the external pressure was attributed to the reduction of the magnitude of magnetization due to the alteration of the AFM/FM magnetic interaction.

3.3 Magnetocaloric properties in Ni-Mn-Ga alloys in various reduced dimensions

The magnetocaloric properties of the alloys in reduced dimensions such as ribbons, microwires and thin films are of considerable interest as they possess a high surface area to volume ratio, high porosity, etc [71-74] that affects the magnetic and magnetocaloric property differently as compared to their bulk counterpart. The magnetic and magnetocaloric properties of the alloys in the form of ribbons, microwires and thin films have been overviewed in the following sections.

3.3.1 Magnetocaloric properties in Ni-Mn-Ga alloys in the form of ribbons

Ribbons of these alloys can be prepared by a well-known melt-spun technique. This is a cost-effective method that can produce ribbons of these alloys with a high cooling rate of 104 –105 K/s. Early investigation of Ni-Mn-Ga melt-spun ribbons revealed that they exhibit some specific properties compared to bulk alloys [75, 76]. Rapid quenching from the liquid state strongly influences the alloy's martensitic structure and microstructure, which in turn affects the characteristic transition temperatures as well as the magnetic property of the alloys. An increase in quenching rate results in the decrease of the characteristic temperature such as ferro-para magnetic, martensitic, and premartensitic transformation temperatures. It also decreases the saturation magnetization of both the phases. This effect was ascribed to short-range chemical disorders within sublattices and the introduction of some internal stress [23, 77]. By subsequent annealing, the internal stress can be reduced and the ordering can be improved of the melt-spun ribbons. Consequently, increases all the mentioned characteristics after heat treatment of the parent phase. Magnetization anomalies and peculiarities in magnetic properties were also observed in rapidly quenched melt-spun ribbons of these alloys due to concentration inhomogeneities and atomic disorder [78].

There are a significant number of studies have been reported regarding the magnetocaloric effect in melt-spun Ni-Mn-Ga alloys. However, the reported values of isothermal magnetic entropy changes (ΔS_M) (Estimated from Maxwell's equation) are comparable to that obtained from the bulk materials. For example, a maximum ΔS_M of −10.4 J/kg K was obtained in $Ni_{55}Mn_{19.6}Ga_{25.4}$ ribbon at room temperature for a field change of 2 T at the merged magnetic and structural transition temperature reported by Rao et al [73]. A net refrigeration capacity of 45.5 J/kg with reduced hysteresis loss was also observed by them. Li et al. reported a large ΔS_M of -30.0 J/kg K at the magnetic field change of 5 T in annealed ribbons of $Ni_{52}Mn_{26}Ga_{22}$ with magneto-multistructural transformation [31].

3.3.2 Magnetocaloric properties in Ni-Mn-Ga alloys in the form of microwires

Microwires are a promising form of materials that have attracted immense attention from researchers in recent years. Generally, wires with an average diameter ranging from one to several hundred micrometers are considered microwires. Taylor–Ulitovsky technique is a famous technique that allows one to fabricate very long microwires with amorphous, nanocrystalline, or granular microstructures [79]. The fabrication and investigation of magnetocaloric properties of glass-coated microwires of Ni–Mn–Ga Heusler alloy were started by Varga et al. [80] in 2011. Using Taylor–Ulitovsky method they prepared $Ni_{50.95}Mn_{25.45}Ga_{23.6}$, $Ni_{62}Mn_{23}Ga_{25}$ and $Ni_{65}Mn_9Ga_{26}$ glass-coated microwires with inner and outer diameters ranging from 10 to 44 mm and from 65 to 90 mm, respectively [80-82]. X-Ray diffraction and other structural studies revealed that as prepared microwires consist of mixed structures, cubic austenite and tetragonal martensite structure. However, after annealing the microwires become single phase. Annealing makes considerable changes in the sample's structure and induces internal stress by the glass coating [83, 84]. Therefore, only after annealing above 773K for a few minutes, the as-prepared microwires show ferromagnetic ordering and a ferro-para Curie transition appears within 300 - 315 K. Ferromagnetic Heusler alloys in the form of microwires have shown excellent magnetocaloric properties comparable or even higher to those of bulk materials [85, 86]. Reduced hysteresis and a large working temperature interval were achieved by these microwires. Cu-doped Ni-Mn-Ga microwires with diameters of 20–80 μm and lengths of 30–150 mm were fabricated by Zhang et al. using the melt-extraction method [87] Utilizing the magnetostructural coupling and a wide range of martensitic transformation temperatures a high refrigeration capacity of ~78 J/kg at a magnetic field of 5 T was produced in the annealed $Ni_{49.4}Mn_{26.1}Ga_{20.8}Cu_{3.7}$ microwires. Recently, Ni-Mn-Ga microwires with a diameter of 35–80 μm were prepared by Qian et al. using a melt-extraction technique [15]. Negligible hysteresis and relatively good mechanical stability were found due to the high specific surface area (SSA) that reduces incompatibility between neighbouring grains. Owing to the high SSA of these Ni-Mn-Ga microwires, the transformation temperatures (structural and magnetic transformation) were adjusted by the element evaporation at high temperatures. A large tunable MCE was achieved due to coupled

magnetostructural transition attained by proper composition design and subsequent tuning (by element evaporation). A maximum isothermal magnetic entropy change (ΔS_M) of −11.3 and −18.5 J/ kg K were obtained at magnetic fields of 2 and 5 T, respectively. The obtained ΔS_M is comparable to that of polycrystalline Ni-Mn-Ga ribbons, films and bulk alloys. Further, the annealed microwires showed wide working temperature interval (WTI), low hysteresis loss, high heat exchange efficiency along with good mechanical stability. Very recently, a huge refrigeration capacity of ~255 J/kg at a field of 5 T with a large working temperature interval of ~71 K was achieved in bamboo-grained Ni-Mn-Ga microwires [88]. This was achieved by partially coupled magnetostructural transition and a wide temperature range of martensitic transition. Further, almost no hysteresis loss was observed in these bamboo-grained microwires.

3.3.3 Magnetocaloric properties in Ni-Mn-Ga alloys in the form of thin films

The magnetocaloric effects in thin film have been investigated extensively for their potential application of cooling in (nano)microdevices. The deposition of alloy thin film maintaining the desired composition resembling the bulk material is challenging. Moreover, the measurement of the magnetocaloric effect of this alloy film is also difficult as the mass of the material is very small and hard to quantify. Beside all these difficulties a significant number of studies on Ni-Mn-based Heusler alloy thin films have been reported in the literature [89-95]. In these reports, magnetron sputtering, ion beam sputtering and pulsed laser deposition technique have been widely used to deposit thin films of desired stoichiometric and off-stoichiometric crystalline Ni-Mn-Ga alloys with good magnetic and magnetocaloric properties [89-96]. Various substrates were used to deposit thin films of the Ni-Mn-based alloy such as Si, SiO_2, MgO, Al_2O_3 etc. [96-99]. Deposited films show crystallinity however few reports claim annealing is required to get crystalline samples with ferromagnetic order [90]. The critical transition temperatures of these alloys are found to be similar to that of the bulk materials. However, higher stresses and disorder in thin film modify the magnetic property and nature of transition differently. The maximum isothermal magnetic entropy changes in thin film of the Ni–Mn–Ga ferromagnetic shape memory alloys have mostly been reported for compositions with coupled magneto-structural phase transitions [100, 101]. Recarte et al. [98] deposited Ni–Mn–Ga thin film with a thickness of 0.4 μm on Al_2O_3 substrate by RF magnetron sputtering technique. The magnetic and structural transition temperatures in these thin films of composition $Ni_{53.5}Mn_{23.8}Ga_{22.7}$ were 346 K and 343 K respectively. Using Maxwell's relation, a maximum entropy change ΔS_M=−8.5 J/kg K was achieved at a temperature very close to the martensitic transition temperature with a field of 6 T which is relatively small compared to that obtained from the bulk alloy. A few years later another group of researchers [100] deposited $Ni_{51}Mn_{29}Ga_{20}$ thin film of thickness 250 nm using pulsed laser deposition technique. The film obtained with this technique showed good crystallinity as observed from three-dimensional x-ray diffraction data. As the e/a value of this composition (7.73) satisfy the coincidence condition of structural and magnetic transitions for the bulk Ni-Mn-Ga alloy, in the case of thin film it is also observed. A maximum isothermal magnetic entropy change of -1.4 J/kg K for a field change of only 0.5 T was achieved at the coupled structural and magnetic transition temperature.

4. Conclusion and future scope

Ni-Mn-Ga Heusler alloys are promising magnetocaloric material as it displays a large change of magnetic entropy, ΔS_M in bulk material as well as in various form of the alloys like ribbons, microwires and thin films. The first-order magnetic transition associated with the tetragonal to a cubic structural phase change (Martensitic transition) and the second-order magnetic transition (ferro-para Curie transition) are the most important characteristic of this alloy system as all the functional properties including MCE are linked with those transitions. With a fine adjustment of the compositions and/or substitution of the suitable element in this alloy system, these transitions can be tuned up to their coincidence. This deliberate coupling of these first and second-order magnetic transitions is an enhancement criterion to achieve a large/giant magnetocaloric effect. Considering this criterion, a large number of efforts have been made for ribbons, microwires and thin films of these materials also. From these studies, it became obvious that the maximum change of entropy is achieved at the concurrent magnetostructural and Curie transition. Doping/substitution with suitable atoms in this alloy system can modify the structural transition temperature and enhance the magnetocaloric effect. The doping element interacts differently with the other atoms in the austenite and martensite phases and eventually gives rise to a high value of saturation magnetization difference between these phases that led to a giant MCE. There is no significant effect of hydrostatic pressure on magnetic entropy change in these alloy systems, rather it decreases with the increase of pressure.

Comparing the magnetocaloric effect in bulk, ribbons, microwires and thin films of Ni-Mn-Ga alloys it appears that the thin films exhibit much less MCE compared to others whereas the bulk material shows maximum. Ribbons and microwires of these alloys display MCE comparable to that of the bulk alloys along with better mechanical stability and wide working temperature interval. Furthermore, microwires of this alloy favor the element evaporation (Mn) during high-temperature annealing that widens the working temperature interval and reduces hysteresis loss that makes them practically important magnetocaloric material. However, nowadays magnetocaloric materials in the form of thin films are highly demanding for feasible applications in refrigerator bodies of functional microsystems, there is not much report regarding the investigation of MCE in thin films of these materials. Maintaining the desired composition of these alloys in thin films is a difficult task. There are several factors involved in the thin film system such as strain due to crystal mismatch, large surface-to-volume ratio, surface spin canting, suppression of magnetic moment at the interface etc. that may reduce the MCE compared to that of bulk materials. However, to confirm the actual contribution of those factors

toward MCE and to get large ΔS_M more investigations are required.

Conflict of Interest
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