1-1-2015

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Recommended Citation

Rodionov, I., Koshkid'Ko, Y., Cwik, J., Quetz, A., Pandey, S., Aryala, A., Dubenko, I., Stadler, S., Ali, N., Titov, I., Blinov, M., Prudnikov, V., Lahderanta, E., Zakharchuk, I., & Granovsky, A. (2015). Peculiarities of Giant Magnetocaloric Effect in Ni50Mn35In15 Alloys in the Vicinity of Martensitic Transition. *Physics Procedia, 75*, 1353-1359. https://doi.org/10.1016/j.phpro.2015.12.151

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Physics Procedia

Volume 75, 2015, Pages 1353–1359

20th International Conference on Magnetism

Peculiarities of giant magnetocaloric effect in $Ni_{50}Mn_{35}In_{15}$ alloys in the vicinity of martensitic transition

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Abstract

Adiabatic temperature change, resulting from MCE, is a complex effect depending on in-field behavior of both lattice and magnetic subsystem of the sample. For studying MCE, the understanding of their interplay is crucial. Comparing ΔT_{ad} in different fields (e.g. below, above and far above saturation field) it should be possible to derive contributions from lattice and magnetic subsystems. In this present work we present our study of the MCE in $Ni_{50}Mn_{35}In_{15}$ Heusler alloy in the vicinity of the magnetostructural transition in 1.6 T and 14 T field.

Keywords: Heusler alloys, magnetocaloric effect, martensitic transition,magnetic fields

Recent studies of magnetic refrigeration propose materials undergoing 1^{nd} and 2^{nd} order transitions as a refrigerants because of large entropy and adiabatic change of temperature ΔT_{ad} exhibited by these materials on applying the magnetic field[1, 2]. The development of this topic requires better understanding of the in-field behavior of both lattice and magnetic subsystem of the material as they contribute to ΔT_{ad}[3, 4]. NiMn-based Heusler alloys undergoing 1^{st} order magnetostructural martensitic transition around room temperature are one of the proposed materials for magnetic refrigeration[5, 6, 7]. In these materials the transition can be induced by both temperature and field change, influencing on both lattice and magnetic subsystems[8]. Effect of the field application in the vicinity of the transitional hysteresis depends on the shape of the structural hysteresis: e.g. if the field-induced temperature shift of the hysteresis is larger than the width of the hysteresis itself one will observe magnetic field induced structural transition[9]. These ideas were later applied to analysis of the influence of the transition hysteresis on measured ΔT_{ad} on the initial application of the field and following field cycling[10]. It was shown that ΔT^{ad} depends on whether the initial measurement temperature

Selection and peer-review under responsibility of the Scientific Programme Committee of ICM 2015

doi:10.1016/j.phpro.2015.12.151

lies on the forward or on the reverse transformation path of the thermal hysteresis and whether the field applied initially or the field cycling is performed. To extend the knowledge of the field behavior of ΔT_{ad} one should consider the *extreme* case of fields far above the saturation field as these fields should be enough both to induce structural transition and saturate the sample. Certain approach was performed by novel optical imaging method[11]. The H-T phase diagram for Ni-Co-Mn-In sample was studied using this method. However due to the technical issues the ΔT_{ad} was measured only in Gd sample. Effect of the 14 T field cycling on ΔT_{ad} in Ni-Mn-In-Co sample was studied in [12]. The direct measurements performed at initial temperature 273 K gave the result similar to observed in Ni-Mn-In sample in[10]: reversible temperature change on applying magnetic field after first cycle. However, this work should be extended by temperature dependent ΔT_{ad} measurements. In this work we present the results of our study of ΔT_{ad} in ternary $Ni_{50}Mn_{35}In_{15}$ Heusler alloy on application of 1.6 T and 14 T fields in the vicinity of the martensitic transition in temperature range between 300 and 350 K and relate them to the behaviour of magnetic and lattice subsystems following the models published in [9, 10] and extend them to high field case.

Polycrystalline ingot was prepared by arc-melting pure metals (Ni, Mn: 99.9%, In: 99.9995%) in argon atmosphere, then the ingot was annealed at 1123 K in vacuum ($\sim 1.3 \cdot 10^{-7}$ bar) for 24 hours. Finally, the ingot was tempered. The composition of the alloy was determined by energy dispersive X-ray spectroscopy. The following measurements presented in this work were performed on the samples cut from the same ingot. Magnetization measurements in fields up to 1.6 T were performed using vibrating sample magnetometer (Lake Shore VSM). Adiabatic temperature change ΔT_{ad} in fields up to 1.8 T was measured using MagEq MMS 801 setup. The rapid field change was performed by rotating permanent magnets at 2 T/s rate. ΔT_{ad} in fields up to 14 T was measured by means of home-assembled setup developed by Yu. S. Koshkid'ko and J. Cwik based in International Laboratory of High Magnetic Fields and Low Temperatures, Wroclaw. The schematic of this setup is shown in Figure 1. The dewar flask is fixed inside the Bitter magnet. The area with homogeneous field is about 5 cm long. Thermocouple and heater for controlling the temperature are mounted inside the flask. The sample holder consists of the long shaft and the sample chamber, where the sample, type **T** thermocouple for measuring the sample temperature and Hall sensor for the field measurement are mounted. Applying the field is performed by inserting the sample space from region with zero field into the Bitter magnet. The achieved field change rate is about 5.6 T/s in this case. In both cases ΔT_{ad} change was measured consequently on both heating and cooling: after stabilizing the measurement temperature, magnetic field was applied and removed, and the sample was brought to the next measurement temperature.

Figure 2 shows the temperature dependence of the magnetization measured in 5 mT. The measurement was performed in three temperature sweeps: warming the sample in field from the demagnetized state (ZFC), cooling in field (FC) and warming the sample again (FW). The temperature dependence of magnetization has following features: the splitting between ZFC and FW curves (attributed to the presence of short-range AF interactions in martensite state [13]), Curie temperature of martensite $T_C^M = 180$ K and the temperature hysteresis due to martensitic transformation, starting at $M_S = 319$ K and followed by Curie temperature of austenite at $T_C^A = 328$ K.

In Figure 3a we show the temperature dependence of magnetization in 1.6 T measured on heating and cooling in the vicinity of the magnetostructural transition. Temperature dependence of magnetization exhibits a hysteresis due to martensitic transition, decrease of magnetization above the transition is related to approaching T_C^A. Note that due to vicinity of M_S to T_C^A maximum magnetization during the transition is about 40 times higher compared to 5 mT case.

In Figure 3b we show ΔT_{ad} on the applying 1.6 T magnetic field on the reverse (heating) and direct (cooling) transformation branches. Initially, at 300 K ΔT_{ad} is insignificant, close to zero, on further heating, at 310 K, it starts decreasing until the minimum of -0.8 K at 320 K. On further heating it starts increasing and reaches maximum of +0.8 K at 324 K, then it decreases, remaining positive. On cooling from above martensite start M_S temperature ΔT_{ad} increases towards maximum of +1.1 K at 321 K, on further cooling it decreases down to almost 0 below 314 K. The splitting between conventional ΔT_{ad} on heating and cooling is 4 K.

Figure 4 shows ΔT_{ad} on the applying 14 T magnetic field on the reverse (heating) and direct (cooling) transformation branches. The inset plot covers whole measured temperature range, while the main figure has the same scale as in Figure 3 for ease of comparison. At 250 K ΔT_{ad} is about 1 K, on further heating it decreases down to 0 at 280 K, and reaches its minimum of -11 K at 314 K, then is starts increasing, reaches 0 at 321 K, continues increasing and reaches maximum of +7 K at 325 K, on further heating it starts decreasing. On cooling ΔT_{ad} shows the same behavior as on heating, although it exhibits maximum of +8 K at 320 K, reaches 0 at 318 K and exhibits minimum of -10 K at 310 K. The splitting between ΔT_{ad} on heating and cooling is 2.4 K.

As it was mentioned above, lattice and magnetic subsystems of the sample provide different impact on ΔT_{ad}: the adiabatic magnetization of ferromagnet results in positive temperature change, while lattice transformation results in negative temperature change in NiMn-based Heusler alloys. When the 1.6 T field is applied to the system residing on the reverse transformation branch (i.e. the branch on warming from martensite to austenite phase) negative ΔT_{ad} is observed. This happens because applied magnetic field both orders the magnetic moments and induces growth of the austenite phase. The resulting sum of these impacts is negative. When the 1.6 T field is applied to the system residing on the direct transformation branch positive ΔT_{ad} is observed. That is due to the fact, that this field orders magnetic moments, but is not enough to induce martensitic transition in any direction. One can conclude this from the fact that ΔT_{ad} is nonnegative function between 300 K and 350 K. When the 14 T field is applied to the system residing on the reverse transformation branch ΔT_{ad} is negative, however, the temperature region with negative ΔT_{ad} is much larger in this case: from 280 K to 321 K in this case vs. from 311 K to 321 K in 1.6 T. This happens because with 14 T field austenite phase can be induced at lower temperatures compared to 1.6 T case. However, on the direct transformation branch we also observe negative temperature change between 280 K to 318 K, what is different from lower field case. This happens because with 14 T field it is possible to induce austenite phase even on the direct transformation branch. 14 T field is high enough to induce the full transformation from given temperature above A_S (i.e. from certain austenite-to-martensite ratio state to 100% austenite phase). Additionally this explains the small difference temperature hysteresis between MCE on heating and cooling in 14T field: main impact to MCE in this case is negative ΔT_{ad} arising from induced structural change, positive impact from magnetic subsystem is small as it is driven far beyond saturation with these fields, however, in the end of the reverse transformation branch and in the beginning of direct transformation branch its impact is comparable to impact from structural change.

In this work we have studied the effect of applying different magnetic fields to NiMn-based Heusler alloy in the vicinity of a magnetostructural transition. Applying 1.6 T field resulted in inverse MCE on the reverse transformation branch with maximum effect of 0.8 K, and conventional MCE on the direct transformation branch. We relate this behavior to the fact that applying the field induces structural changes on the reversal transformation branch, while on the direct transformation ranch it does not. Applying 14 T field to the system on the reverse transformation branch results in inverse MCE with maximum effect of 11 K. This behavior is similar to the behavior in 1.6 T, however the magnitude of the effect is much larger. Applying 14 T leads to different behavior on the direct transformation branch compared to 1.6 T: we observe inverse MCE in this case. We relate this behavior to the fact that applying 14 T field induces structural change on both branches of the martensitic transition.

Authors would like to acknowledge the financial support by European Stoical Foundation (Grant No 15-02-02077 and 15-52-78080), by the Office of Basic Energy Sciences, Material Science Division of the U.S. Department of Energy (DOE Grant No. DE-FG02-06ER46291 and DE-FG02-13ER46946) and by Russian Foundation for Basic Research (Grant No 15-02-01976).

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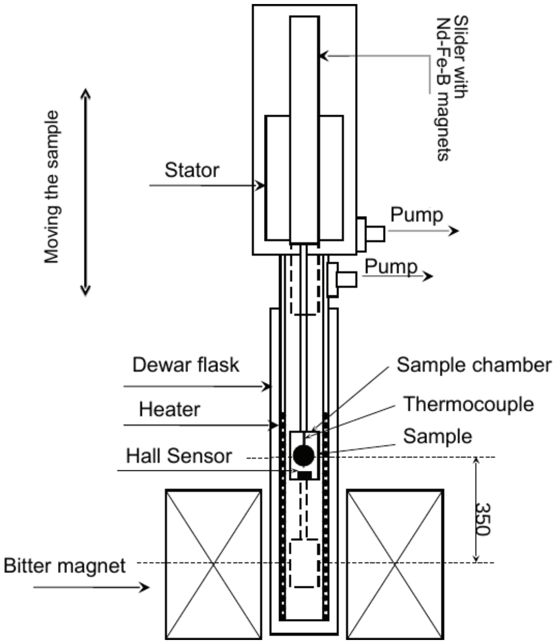

Figure 1: Schematic overview of high field setup for ΔT_{ad} measurements developed by Yu. S. Koshkid'ko and J. Cwik.

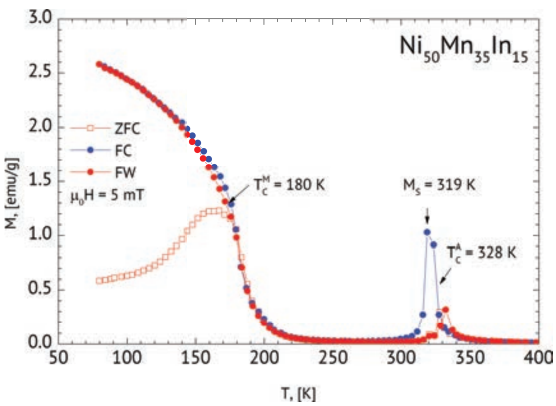

Figure 2: Temperate dependence of magnetization in 5 mT field for $Ni_{50}Mn_{35}In_{15}$ sample.

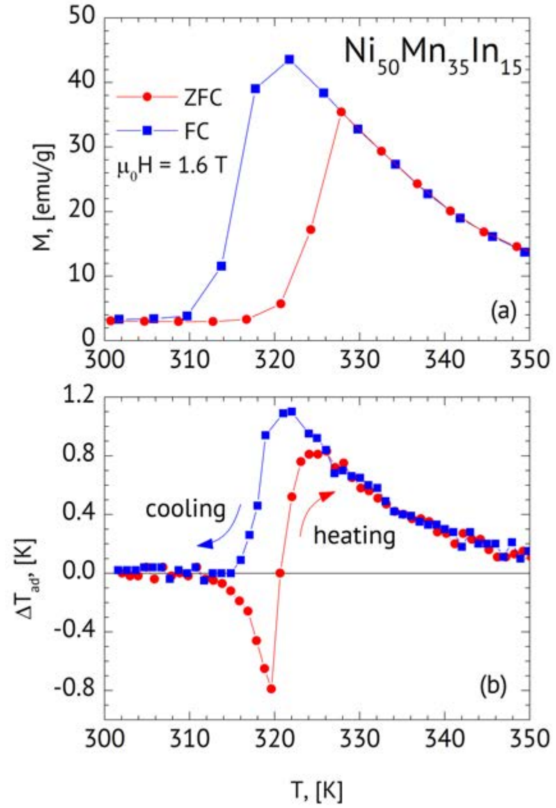

Figure 3: Temperate dependence of magnetization and ΔT_{ad} in 1.6 T field in the vicinity of martensitic transition for $Ni_{50}Mn_{35}In_{15}$ sample.

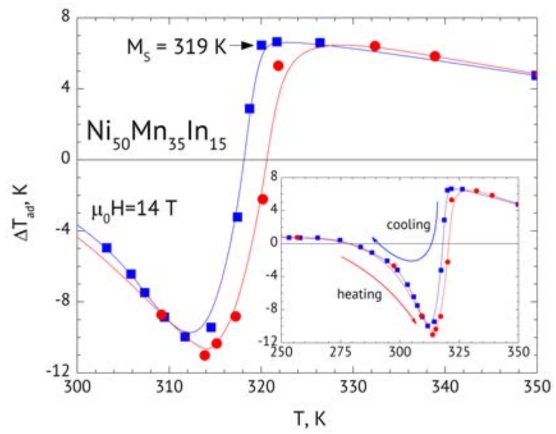

Figure 4: Temperate dependence of magnetization and ΔT_{ad} in 14 T field in the vicinity of martensitic transition for $Ni_{50}Mn_{35}In_{15}$ sample.