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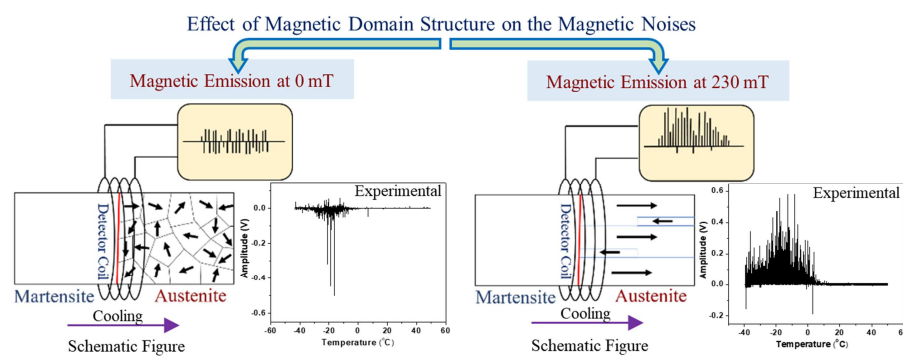
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Thermal, acoustic and magnetic noises emitted during martensitic transformation in single crystalline $\text{Ni}_{45}\text{Co}_5\text{Mn}_{36.6}\text{In}_{13.4}$ meta-magnetic shape memory alloy

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ABSTRACT

Meta-magnetic shape memory alloys (MMSMAs) display an abrupt change in magnetization across the martensitic transformation. Simultaneous detection and statistical analysis of the emitted acoustic and magnetic noises, accompanied with martensitic transformations are important for better understanding of the magneto-structural coupling, and for utilization of well-characterized acoustic and magnetic signals for practical applications. In the present work, characteristics of thermal, acoustic and magnetic noises, detected during martensitic transformation in the $\text{Ni}_{45}\text{Co}_5\text{Mn}_{36.6}\text{In}_{13.4}$ MMSMA single crystals, were investigated for the first time. Statistical analysis of the thermal spikes, appeared during the calorimetric measurements at low cooling rates (typically below 1 K/min) showed that the energy distributions followed a power law with $\varepsilon=2.0\pm0.3$ exponent, which is in good agreement with the exponents of energy distribution of acoustic and magnetic emission events ($\varepsilon=1.7\pm0.1$ and $\varepsilon=1.7\pm0.2$, respectively). The above exponents were independent of the external magnetic field between $B=0$ and 250 mT. Interestingly, the acoustic and magnetic exponents during cooling and heating were the same in contrast to the expected asymmetry based on the fact that the transformation demonstrates an isothermal character during cooling and an athermal one for heating. Upon interrupting cooling, and dwelling at a temperature above the martensite finish temperature, practically no acoustic and magnetic noises were detected, while upon continued cooling the noise appeared again, with the same critical exponents. This indicates that the main discontinuous events, leading to noise emissions, should be the same before and after dwelling as well as during cooling and heating. In magnetic noise measurements, under zero external field, the magnetic noise packets contained voltage peaks in the detector coils in both (positive and negative) directions. Under small external magnetic fields (between 20 and 230 mT), the activity of magnetic signals increased and the magnetic peaks, during both heating and cooling, appeared more and more unidirectional (i.e. peaks only in one direction were observed) which can be attributed to the increased magnetization of the austenite.

Keywords: Magnetic Noise; Metamagnetic Phase Transition; Shape Memory Alloys; Ferromagnetic Shape Memory Alloys

1. Introduction

Ferromagnetic shape memory alloys (FSMAs) have attracted much attention in the past few decades as high-performance actuator materials due to the large magnetic-field-induced strain (MFIS) resulting from the rearrangement of twin variants in the martensite phases. They are multifunctional materials which can be used in applications such as actuation, sensing, energy harvesting, and magnetic refrigeration [1-3]. They are also capable of generating large amount of shape change under magnetic fields [4-7].

Ni-Co-Mn-In system is a special type of FSMAs where the first order structural and the second order magnetic phase transitions overlap. Thus, these alloys exhibit metamagnetic phase transition and called meta-magnetic SMAs (MMSMAs) [8-10], in which there is a large difference between the saturation magnetizations of ferromagnetic austenite and paramagnetic (or antiferromagnetic) martensite phases. As a result of this unique magneto-structural coupling, MMSMAs demonstrate high magnetic work output levels, nearly perfect magnetoelasticity and magnetic shape memory effect [11-17]. They can also be used in magnetocaloric applications [8, 18, 19, 20, 21, 22, and 23].

During martensitic transformations, the motion of phase boundaries is usually not smooth, but has a stop-and-go character [24-29], regardless of whether the transformation is triggered by thermal, mechanical or magnetic stimuli. Similarly, the stress or magnetic field induced rearrangements of martensite variants demonstrate a jerky character [30, 31] too. Therefore, it is possible to detect both acoustic and magnetic noises, when the martensitic transformation or martensite variant reorientation is accompanied with abrupt changes in magnetization [24, 31].

The nature of the aforementioned jerky character is currently an active area of discussion [24, 29, 32, and 33]. The formation of martensite variants during cooling (or their disappearance upon heating) is an intermittent process characterized by avalanches, which can be described by the power law distribution equation:

$$P(X) \cong X^{-\varepsilon} \exp(-X/X_c), \quad (1)$$

where X can represent the amplitude, size, energy or duration of the jerks, ε is the corresponding exponent, and X_c is the cutoff value [24, 32, 33, 34]. This power law behaviour is a characteristic sign of structural processes showing so-called “driven criticality” [24, 32, 33] and it predicts universal exponents, expected to depend only on the symmetry change during the phase transformations (see e.g. [29]). On the other hand, it was observed in selected experiments that these exponents exhibited detectable differences even between heating and cooling cycles through the martensitic transformation [35]. Similarly, the exponents were determined to vary with the microstructural length scales (e.g. in alloys with and without nano-precipitates [24, 36, and 37]). Therefore, the statistical analysis of noises associated with the jerky behaviour and the time correlations of the noise signals (i.e. whether a given signal is fully independent from the previous events or not) may reveal important statistical information on the fundamental physics behind the martensitic transformation, such as the

nature of the magnetoelastic coupling, and on the change of magnetization during martensitic transformation or martensite variant reorientation.

To date, magnetic and acoustic emission measurements were carried out only in Ni_2MnGa [26, 27, 28, 38] as well as in $\text{Ni}_{49}\text{Fe}_{18}\text{Ga}_{27}\text{Co}_6$ (at. %) FSMA single crystals [24, 36, 37], where both the austenite and martensite phases are ferromagnetic. To the best of our knowledge, these kinds of measurements have not been performed to date on MMSMAs, where both magnetic and structural transitions take place simultaneously. Thus, in the present work, we report, for the first time, acoustic and magnetic noise measurements during temperature-induced martensitic transformation between very weakly magnetic (martensite) and ferromagnetic (austenite) phases in $\text{Ni}_{45}\text{Co}_5\text{Mn}_{36.6}\text{In}_{13.4}$ MMSMA single crystals, i.e. during the meta-magnetic transition. It is anticipated that the identification of characteristics of acoustic and magnetic signals during the meta-magnetic martensitic transformation will also have future implications for the structural health monitoring and sensing, where these materials can be utilized as sensory materials.

There are several unanswered questions related to the noisy character and the asymmetry of the forward and reverse martensitic transformations in FSMA [35, 39, and 40]. Two types of asymmetry have been reported so far. In the first type, it is recognized that during athermal martensitic transformations, the noise activities (and the corresponding exponents) for the forward (from austenite to martensite) and reverse transformations are different, and the rationale is based on the different possible ways of relaxations of the elastic strain energy [35]. In the second type, primarily in MMSMAs, another type of asymmetry is manifested: the forward transformation has isothermal character, whereas the reverse one is still athermal [40]. The observed asymmetry of the properties (ac impedance and dc resistivity in polycrystalline samples), in this second type, is attributed to the effect of magnetic ordering: it opposes the forward and promotes the reverse transformation. Therefore, in the present study, the following question is also addressed: how does this type of asymmetry influence the acoustic and magnetic noises during the forward and reverse transformations in single crystalline MMSMAs?

2. Experimental Procedures

$\text{Ni}_{45}\text{Co}_5\text{Mn}_{36.6}\text{In}_{13.4}$ (at.%) was fabricated by vacuum induction melting of elementally pure constituents. Single crystals were then grown from the alloy using the Bridgman method under helium atmosphere. 1.5mm x 1mm x 4cm square rods were electrical discharge machined from the single crystals, with the long axis of the specimen oriented along the [011] direction. These specimens were then wrapped in tantalum foil and placed into a quartz tube. The tube was exposed to high vacuum ($\sim 3.0 \times 10^{-5}$ Torr) for 30 minutes, and was backfilled with ultra-high purity argon before being sealed. The specimens were then heat treated for 24 hours at 900°C, and subsequently quenched in room temperature water, with vigorous stirring, without cracking the quartz tube.

The calorimetric measurements were carried out similar to the procedure described in Ref. [24]. For the noise analysis during the DSC measurements low cooling and heating rates were used because the peaks split into smaller peaks (which is beneficial for the analysis) only if

the temperature ramp was low [26] (typically below 3 K/min). The mass of the examined crystals was 13 mg. The DSC measurements were carried out with a Perkin Elmer DSC 7 device. The background noise was filtered and the calibration was made with the melting point of ice as well as pure indium for each temperature rate. The energy probability distribution functions were created (assuming that the heights of the DSC peaks are proportional to the elementary energy released or absorbed) after 7 cycles of data collection to ensure enough data points for statistical analysis. The time constant of the device was found to be 6 s. This value was taken into account in the calculation of the contributions of the decay of the overlapping peaks in the DSC spectra [24].

The acoustic signals were detected by a Micro-100S (Physical Acoustic Corporation) piezoelectric sensor connected by a steel waveguide to the sample. The setup has a band-pass from 30 KHz to 1 MHz and a 60dB amplifier was used. Acoustic wave packets were recorded (the details of the data acquisition see below) and the method for the evaluation of the statistics of the distribution of amplitude and energy was similar to those used in Refs. [26, 28, 41].

For the measurements of magnetic noise and permeability the following arrangement was used: The magnetic signals were detected by a pick-up coil (300 turns) wound directly on the sample. The coil was about 5 mm long and made of 0.05 mm-diameter copper wire. The setup has a homemade 60-dB amplifier, with 0-200kHz bandwidth. The sample was placed in a heating element and it was closed in He atmosphere inside a stainless steel tube. The whole system was immersed in liquid nitrogen. The signals were recorded using a National Instruments PCI-6111 multifunction data acquisition board, with a 5-MS/s/channel sampling rate. All parts of the setup were grounded precisely to minimize the noises from the PID controller and the electromagnet. Optimal Wiener deconvolution [42] was used to decrease the noise level in the magnetic signals, thereby getting a larger range of avalanche sizes. For simultaneous detection of acoustic and magnetic emission (AE and ME, respectively) signals, the sample was inserted in an aluminium sample holder fixed on a copper cold finger. The cold finger was immersed into liquid nitrogen. The temperature of the sample was regulated by a halogen lamp heater in the sample holder. The sample holder was placed in the air-gap of a Weiss-type electromagnet, producing constant magnetic field. In both arrangements the temperature was measured with a copper-constantan thermocouple, being in close contact with the sample and controlled by a proportional-integral-derivative (PID) controller: it provided linear heating/cooling during the measurements. For simultaneous detection of AE and ME signals, we used the NI PCI-6111 data acquisition board in two-channel mode. The sampling frequency was 500 kHz. The applied cooling/heating rate for this type of measurements was 5 K/min, which was sufficient to reach baseline separation even in the high activity areas.

3. Experimental Results

The magnetic permeability of the sample was measured as a function of temperature, with the help of 1 kHz sinusoidal signals. The permeability is much higher in the austenite phase

than in the martensite, therefore during the phase transformation, there is an abrupt change in the permeability of the samples. The magnetization curve of the austenite was measured by vibrating sample magnetometer (Fig. 1), parallel with the longitudinal axis of the sample.

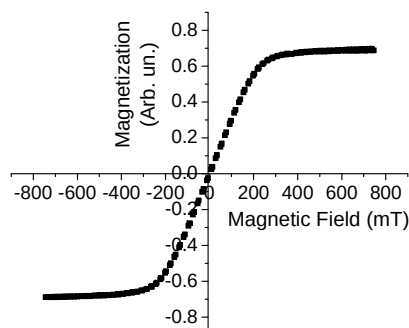
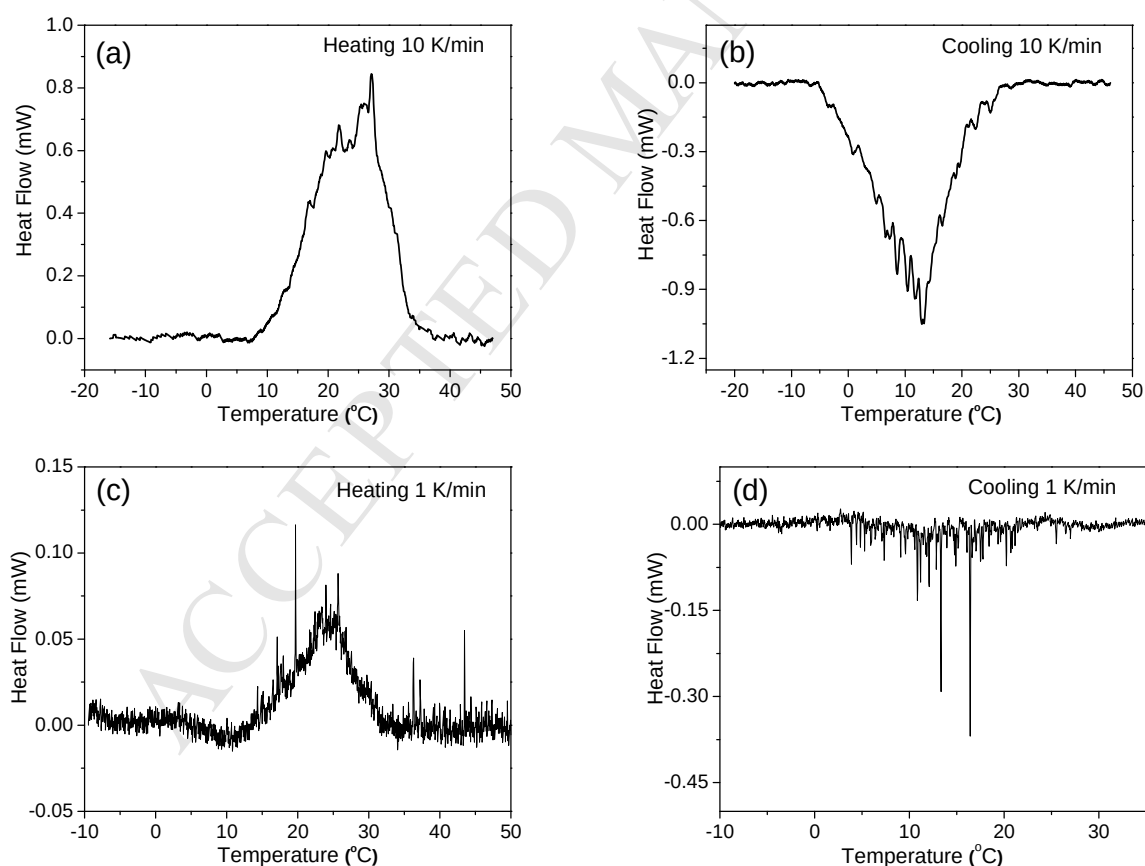


Fig. 1. Magnetic hysteresis loop of the austenite at room temperature: the magnetic field was parallel with the longitudinal axis of the sample (direction [011]).



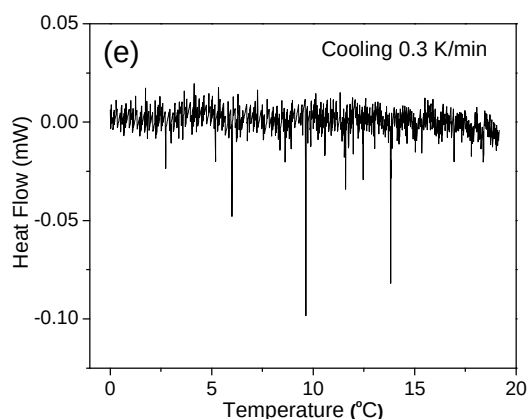


Fig. 2. DSC runs for heating (a, c) and cooling (b, d, e) for the $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals at different temperature rates (10 (a, b), 1 (c, d), and 0.3 K/min (e)).

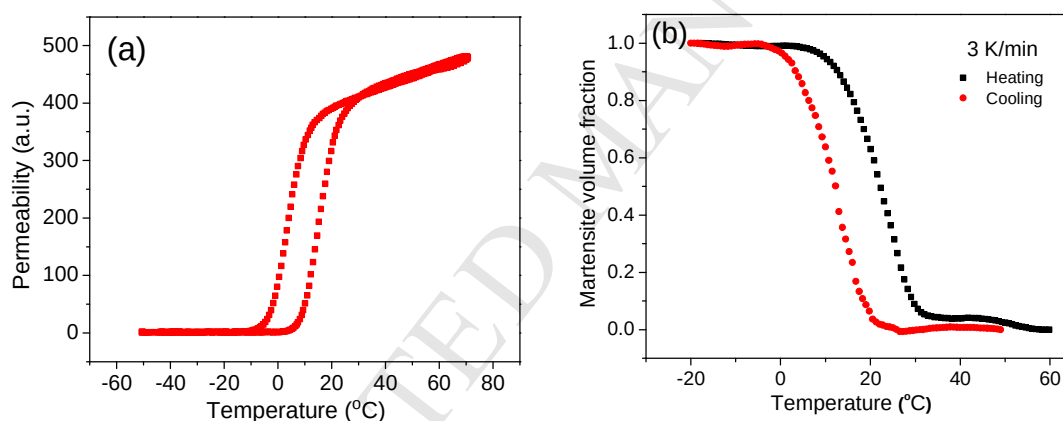


Fig. 3 (a) Hysteresis loop obtained from the permeability measurement (5 K/min, the magnetic field was parallel with the [011] direction). (b) Hysteresis loop calculated from the DSC measurement (3 K/min).

The results of the DSC measurements are shown in Fig. 2. It can be seen that the original continuous envelope curves split into small peaks with decreasing rate, especially for cooling. At the same time the spectra contain smaller and smaller peaks and they are less and less separable from the background: for heating at 0.3 K/min, the peaks were almost at the level of background and this is why it is not shown in Fig.2. It was also possible to construct the hysteresis loops of the martensitic transformation from the measured DSC curves, using the normalized partial integrals of the Q/T curves [43, 44] (Q is the heat of transformation, T is the temperature and the value of the entropy, used for normalization, was determined from the measured DSC curves following [44] and was about 20 J/kgK) for the estimation of the martensite volume fraction, ξ (Fig. 3b). These are in good agreement with the hysteresis loop determined from the permeability measurements (Fig. 3a).

The statistical analysis of the measured split DSC curves was carried out in a similar way as described in Refs. [25, 26], using the proportionality between the peak height and its energy [25]. The background noise was filtered with a properly chosen threshold. The individual peaks in our DSC have relatively long exponential decay, which can lead to overlapping with the next peak(s). Thus, first, the time constant of the DSC device (Perkin Elmer DSC 7) was determined by applying a short laser pulse to the calibration sample in isotherm mode, and from the response of the device the time constant ($\tau=6\text{s}$) was calculated. It was obtained that the peaks at rates higher than 1K/min considerably overlapped, while below 1K/min the overlaps were more moderate and the estimation of the real peak areas could be carried out (i.e. corrections, due to the decay time related to the thermal inertia of the DSC system, were made). Thus the corrected peak values belonging to 0.3 K/min can be accepted as more reliable, and the energy probability distribution function clearly showed a power law behaviour with a cutoff region (Fig. 4a). The relation (1) can be fitted to the experimental distribution functions using a three-parametrical nonlinear fitting by the Levenberg-Marquardt least-squares method. The fitted exponents-were also checked with the maximum likelihood method [45,46]. For the calculation of the exponents, logarithmic data binning was used (the whole interval was split into 15 parts). The maximum-likelihood fitting in Fig. 4b shows that the damping has an influence on the estimation of the energy exponent: this curve is similar to the curve shown in Fig. 4b of [46] for $\Lambda=E_c=10$ (E_c is the cutoff value), with no expressed plateau on it, and thus the value of ε was estimated as the asymptotic value. Thus our error (taking also into account the repeatability and the sensitivity to the cooling rate) is relatively large. It is about ± 0.3 , also because of the narrow energy interval of the available energy values and because the exponent was sensitive to the choice of the threshold.

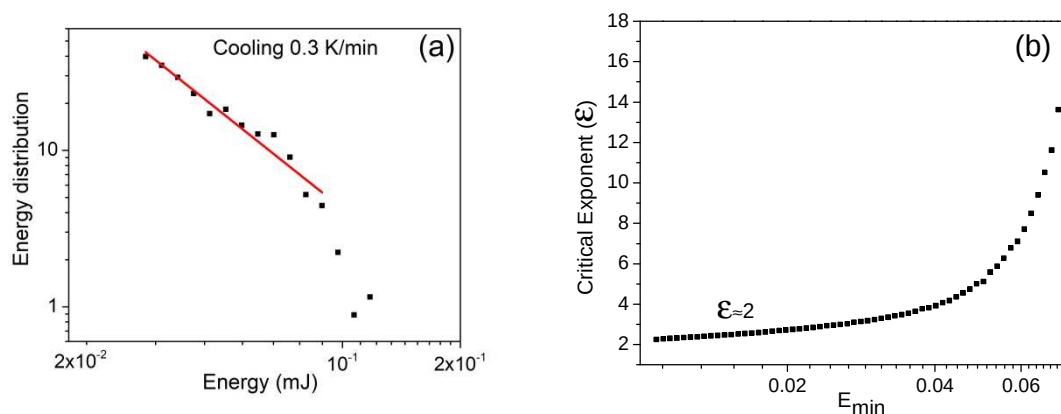


Fig. 4 Log P versus log E for the cooling cycle, according to eqn. (1) (summarized for 7 heating/cooling cycles with 0.3 K/min cooling rate, the number of hits was 3600). The energy units of the horizontal axis are given in mJ from the mW values provided by the DSC, taking $\tau=1\text{ s}$, and the straight line shows the linear part of the curve with $\varepsilon=2.0\pm 0.3$ (a). Maximum likelihood fit; its asymptotic value is about $\varepsilon\approx 2$ (b).

For the measurements of acoustic and magnetic noises, a new sample was used and preliminary temperature runs (in the set up for the simultaneous measurements of AE and ME) indicated a difference in the start and finish temperatures, as compared to the previous samples (this difference can be related to the potential differences in the composition and thermal history). Thus, one DSC run on this new sample, with mass 35 mg, was also performed. The DSC runs for the new sample and the corresponding simultaneously measured acoustic (AE) and magnetic emissions (ME) noises (at 5 K/min rates) are shown in Fig. 5. The AE activity for heating was very low and we could detect only small signals. Thus, in this case the identification of the start and finish was ambiguous (Fig. 5c).

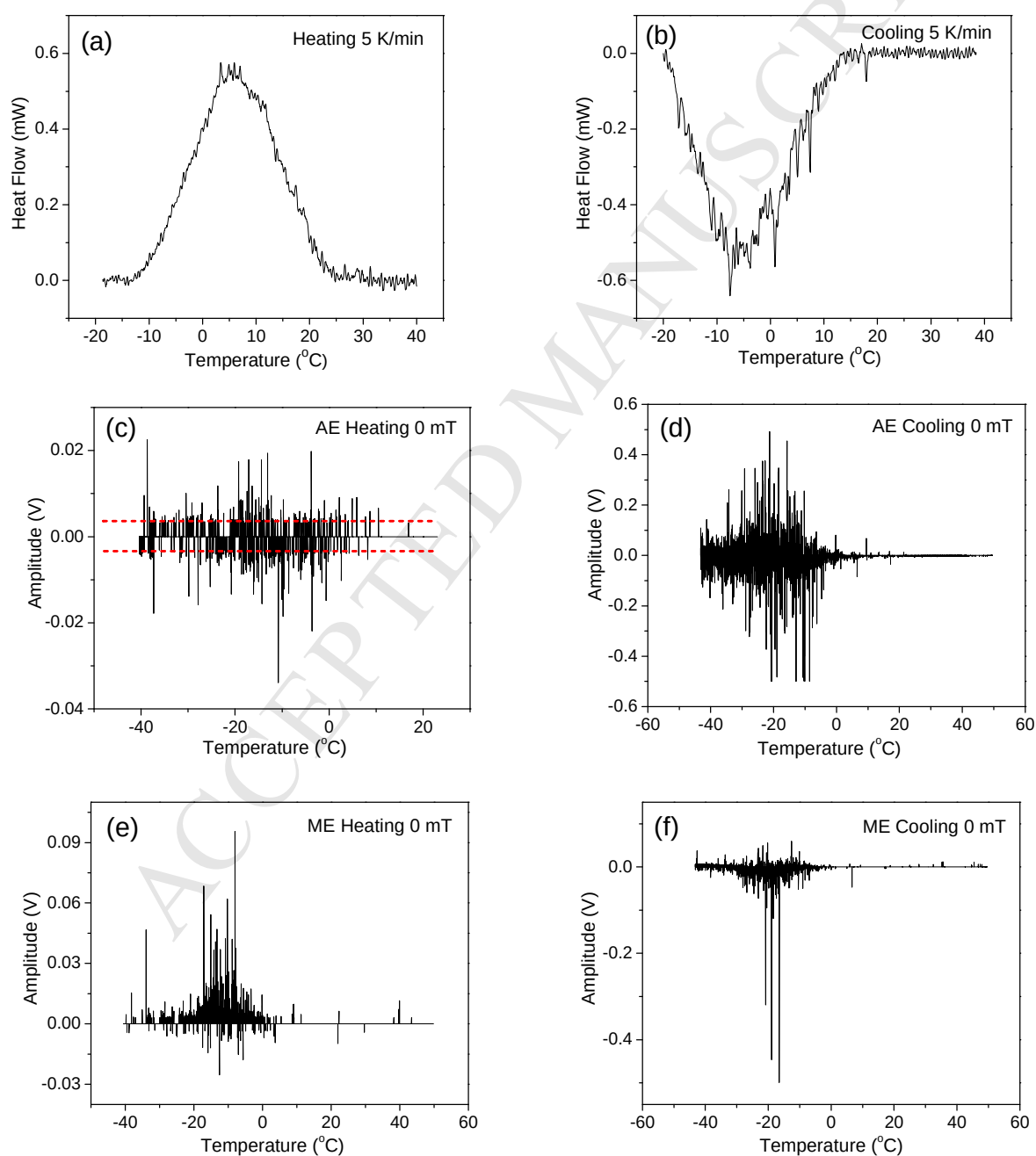


Fig. 5 DSC runs (a, b), acoustic emission (c, d), and magnetic emissions emitted (e, f) during the transformation for heating (left) and cooling (right) for the $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals at the temperature rate of 5 K/min. Voltage peaks in (e, f) belong to magnetization jumps, and horizontal dotted lines in (c) show the threshold levels.

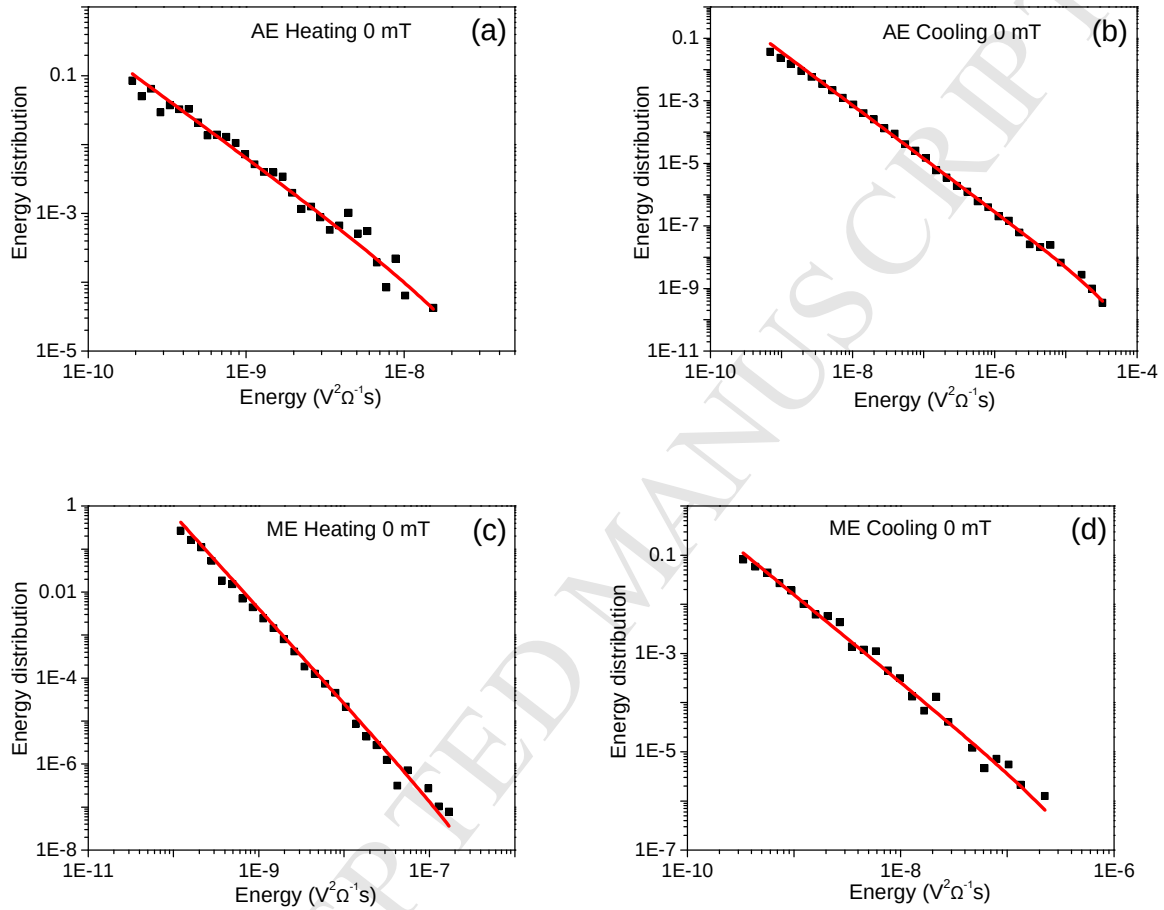


Fig. 6. Energy distribution functions; Log P versus log E, obtained from the acoustic and magnetic measurements during heating (left) and cooling (right) of $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals without an external magnetic field (5 K/min). The numbers of hits were 414 for a), 30063 for b), 12212 for c) and 1097 for d), respectively.

For the determination of the energy probability distribution functions for acoustic emission, the energy of an individual acoustic event, E_i , was determined from an approximate integration, taking the half of the product of the square of the voltage signal with the time (see also [24, 28]). Thus, both the energy and amplitude distributions were determined. Similarly for magnetic signals, the energy of the peak was proportional to the integral of the square of the peak amplitude (see e.g. [28]). In this case, the area of the signals was also calculated according to their usual definition (e.g. the area is proportional to the product of the magnetic

signal amplitude and its duration). Fig. 6 shows, for illustration, the energy distribution functions obtained at zero magnetic field ($B=0$) during heating and cooling for AE and ME. The values of the energy exponents for AE were the same for cooling and heating and $\varepsilon=1.7\pm0.1$. It can be seen that the value of the energy exponent from the DSC measurement agrees well with the value for cooling within the error bar. This is also in agreement with the general rule [25]: since both methods detect the energies of the individual events, their distribution should have the same critical exponents.

Since the signal-to-noise ratio of the magnetic and acoustic noises during the phase transformation without an external magnetic field ($B=0$ mT) was poor (see e.g. Fig. 5c), simultaneous acoustic and magnetic noises were also detected during one cycle of heating and cooling at different small external magnetic fields (between $B=20$ and $B=230$ mT). The magnetic field was parallel with the longitudinal axis of the sample (direction [011]). The results are shown in Figs. 7, 8 and 9.

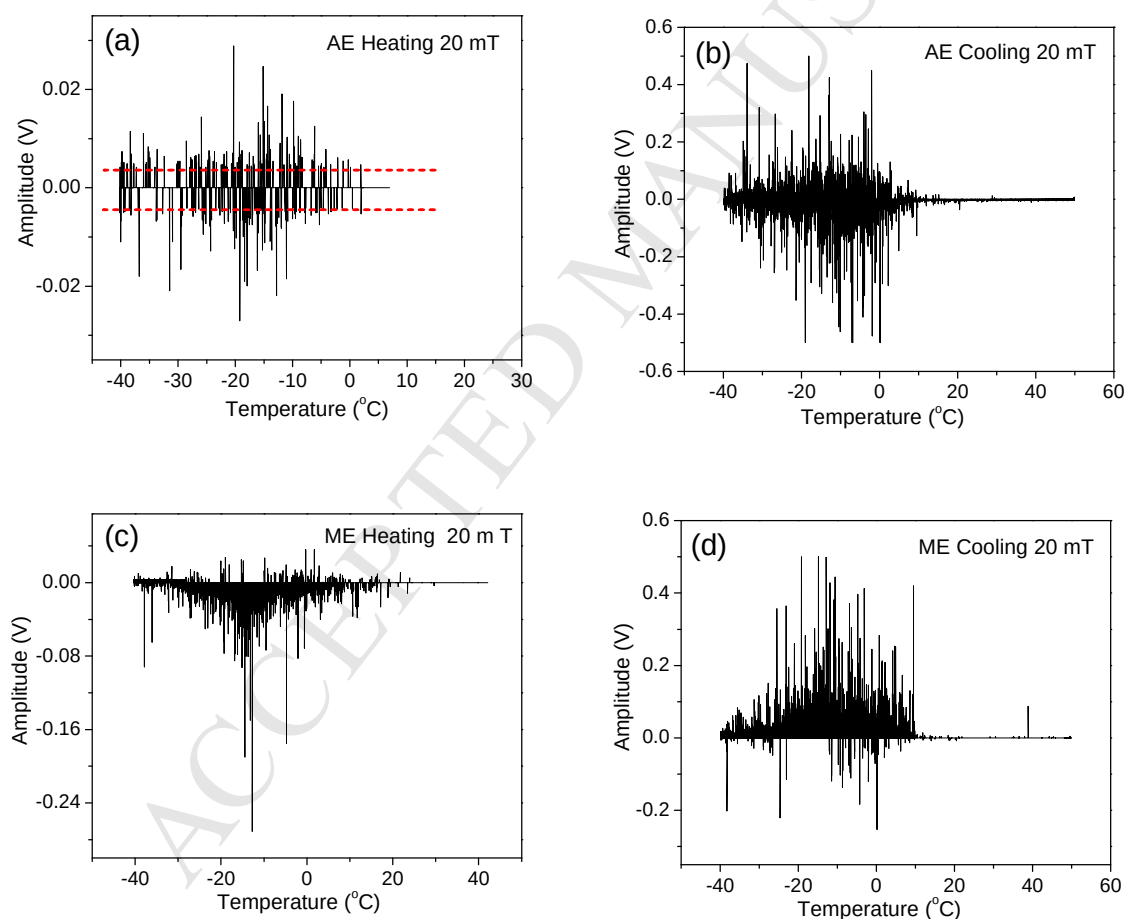


Fig. 7 Acoustic (a, b) and magnetic (c, d) emission signals during the transformation: heating (left) and cooling (right) at $B=20$ mT for $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals.

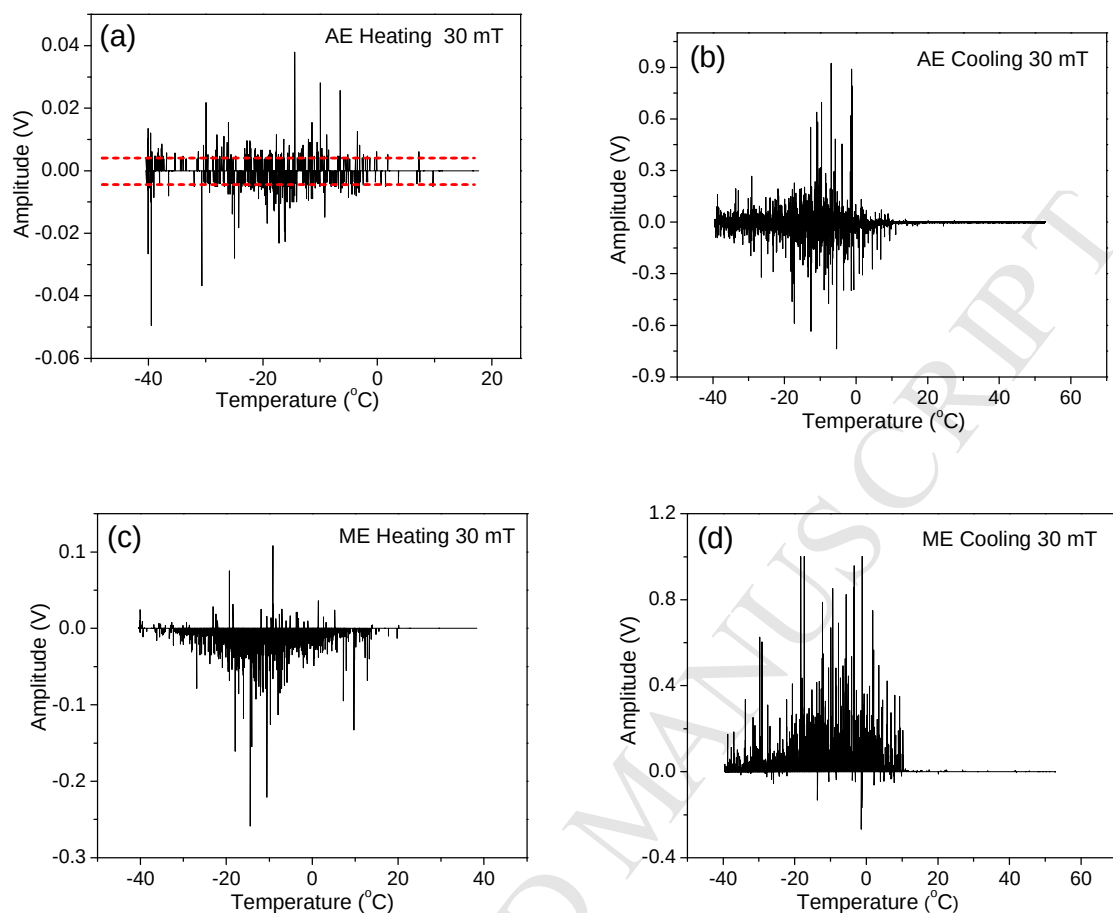
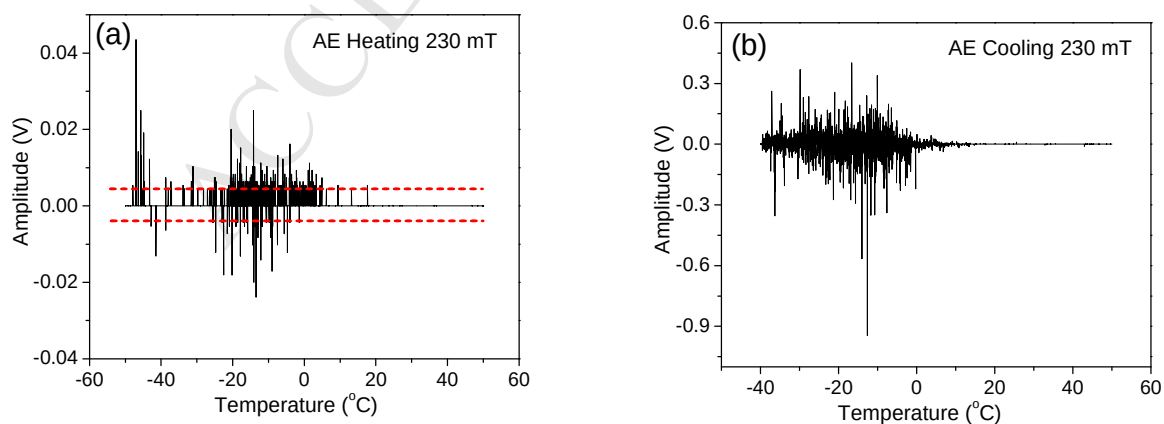


Fig. 8 Acoustic (a, b) and magnetic (c, d) emission signals during the transformation: heating (left) and cooling (right) at $B=30$ mT for $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals.



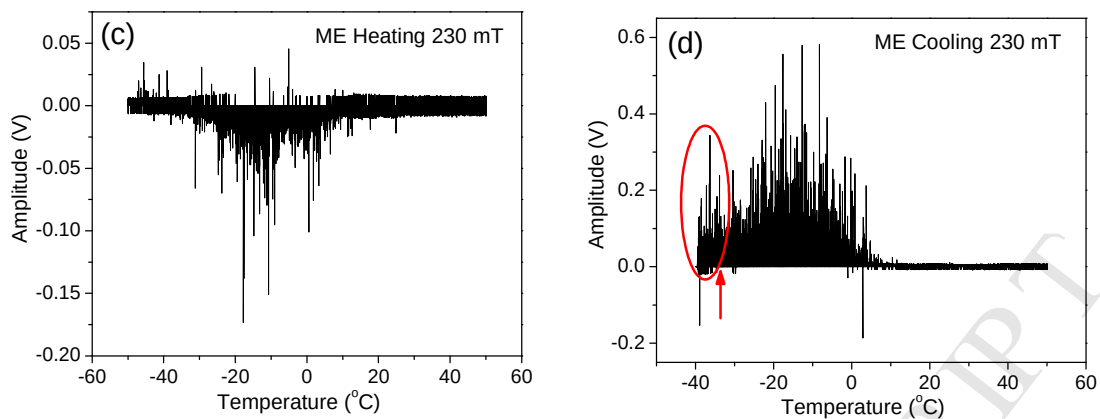


Fig. 9 Acoustic (a, b) and magnetic (c, d) emission signals during the transformation: heating (left) and cooling (right) at $B=230$ mT for $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals. Note the increased activity at low temperatures (area indicated by the arrow), as compared to Fig. 5f.

All of the samples were measured with the same amplification and detection coils, so the observed differences in the magnetic noise intensity are caused by the differences due to the inherent properties of the samples. It can be seen that at $B=0$ mT the peaks are small (Fig. 5, see the scale on the vertical axis) and the whole spectrum is very noisy. At $B=20$ mT (Fig. 7), the peaks are higher and look as typical for similar magnetic noise measured during martensitic transformation [37]. A notable observation is that the peaks for heating as well as cooling are mainly bidirectional (i.e. either positive or negative amplitudes appear) at $B=0$ mT. Increasing the external field, the unidirectional character becomes more and more clear (see the spectra at $B=20$, 30 and 230 mT). Furthermore, from the comparison of magnetic noises at $B=0$ mT and $B=230$, it can be seen that a well-expressed higher activity appears at the end of the cooling part (compare Figs. 5 and 9 for ME with the area shown by the arrow). Nevertheless, the exponents of the energy, amplitude, area and width of magnetic noise peaks in the whole spectrum did not depend on the external magnetic field, within the experimental errors. Table 1 shows these values for magnetic noises for illustration at different external magnetic fields. For comparison, Table 2 presents energy exponents obtained from the acoustic noises. It is worth noting that all our maximum likelihood fits did not provide signs of overlapping different elementary processes. Fig. 10 shows, as an illustration, the maximum likelihood fit for the energy distribution of AE during cooling in $B=30$ mT magnetic field. It can be seen that there is no kink on this curve, which would be expected according to Figs. 2b and 3b of [46], where the kink appeared due to the overlap of two distinct processes with exponents 2.0 and 1.5.

Table 1. Exponents of magnetic noises at different external fields, during thermal cycling at 5 K/min. The error bars are typically ± 0.1 , except for the values at $B=0$ mT, where it is about ± 0.2).

	Energy Exponent		Amplitude		Area		Width	
B (mT)	up	down	up	down	up	down	up	down
0	2.0	1.7	2.1	2.3	2.3	2.2	2.3	2.5
20	1.8	1.7	2.4	2.3	2.3	1.9	2.4	2.2
30	1.6	1.6	2.4	2.2	2.2	1.9	2.5	2.2
230	1.6	1.7	2.1	2.2	2.2	2.2	2.5	2.5

Table 2. Exponents of acoustic noises at different external fields, during thermal cycling at 5 K/min. The error bars are typically ± 0.1 , except for the amplitude exponents, which are ± 0.3 .

	Energy Exponent		Amplitude Exponent	
B (mT)	up	down	up	down
0	1.7	1.7	2.9	2.4
20	1.8	1.7	2.4	2.4
30	1.8	1.7	2.6	2.4
230	1.7	1.7	2.7	2.6

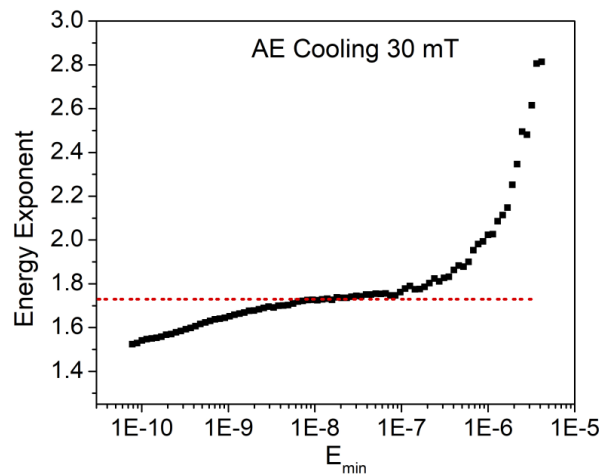


Fig. 10 Maximum likelihood fitting for the estimation of the energy exponent for AE during cooling in B=30 mT magnetic field (see also Fig. 8b).

Regarding the isothermal/athermal character of the transformation, it was shown in [40] that in a polycrystalline alloy with almost the same composition like ours, the austenite to martensite transformation was isothermal, while the reverse one was athermal. Thus, we have carried out the following additional measurements. The cooling was stopped at -34°C (before the M_s would be reached) and the temperature was kept constant for 5 minutes, then the cooling was continued down up to -60°C . Fig. 11 shows the acoustic and magnetic noises versus the time without external magnetic field during these measurements. Practically, no ME during and after dwelling (or only very small voltage peaks) was observed, but a well visible activity appears under the applied magnetic field ($B=30\text{ mT}$) during continued cooling (Fig. 12). Because of the very low magnetic activity, the noises collected before and after the stop temperature were separately evaluated only for the acoustic noises at $B=0\text{ mT}$. The

energy distribution functions, determined separately for these two parts, are shown in Figs. 13 and 14.

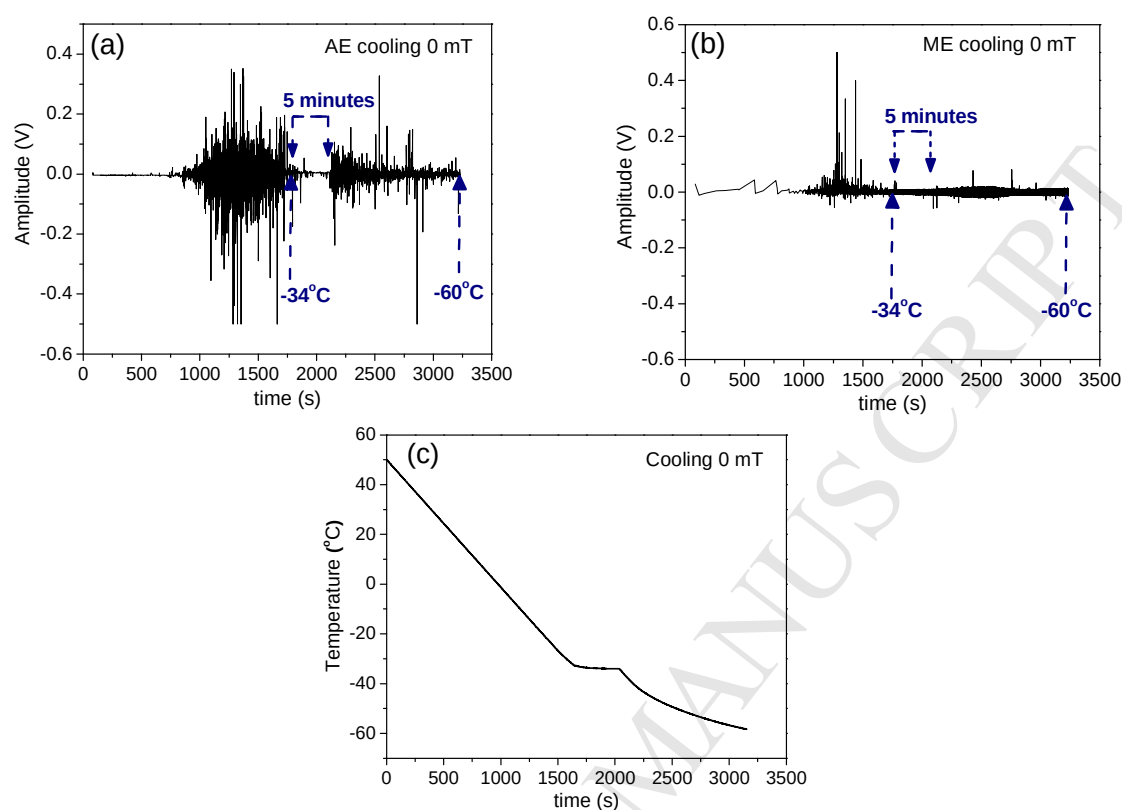


Fig. 11 Acoustic and magnetic noises during cooling (the temperature decreases from left to the right) without external magnetic field in $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals (obtained using a 5 K/min cooling rate).

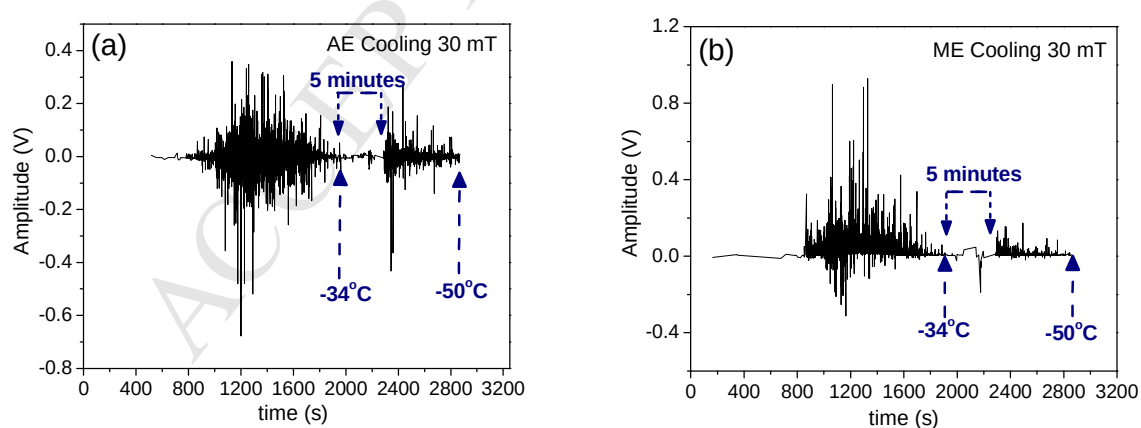


Fig. 12 Acoustic and magnetic noises during cooling under external magnetic field ($B=30$ mT) in $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals (obtained using a 5 K/min cooling rate).

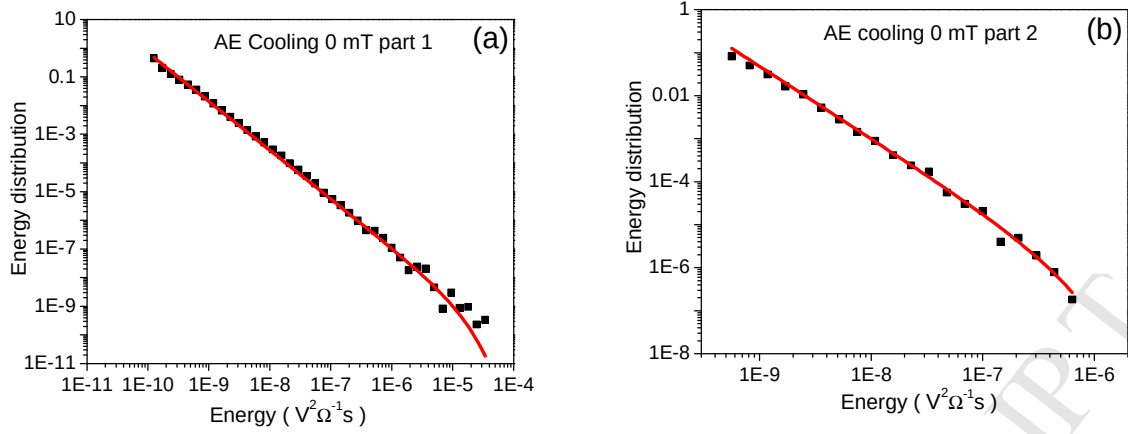


Fig. 13 Energy distribution functions; Log P versus log E, obtained from acoustic measurements for parts 1 and 2 (before and after dwelling, respectively), during cooling in $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals without an external magnetic field. The exponents are the same within the experimental errors for: (a) $\varepsilon=1.7$, and (b) $\varepsilon=1.7$. The numbers of hits were 49791 for a) and 3727 for b), respectively.

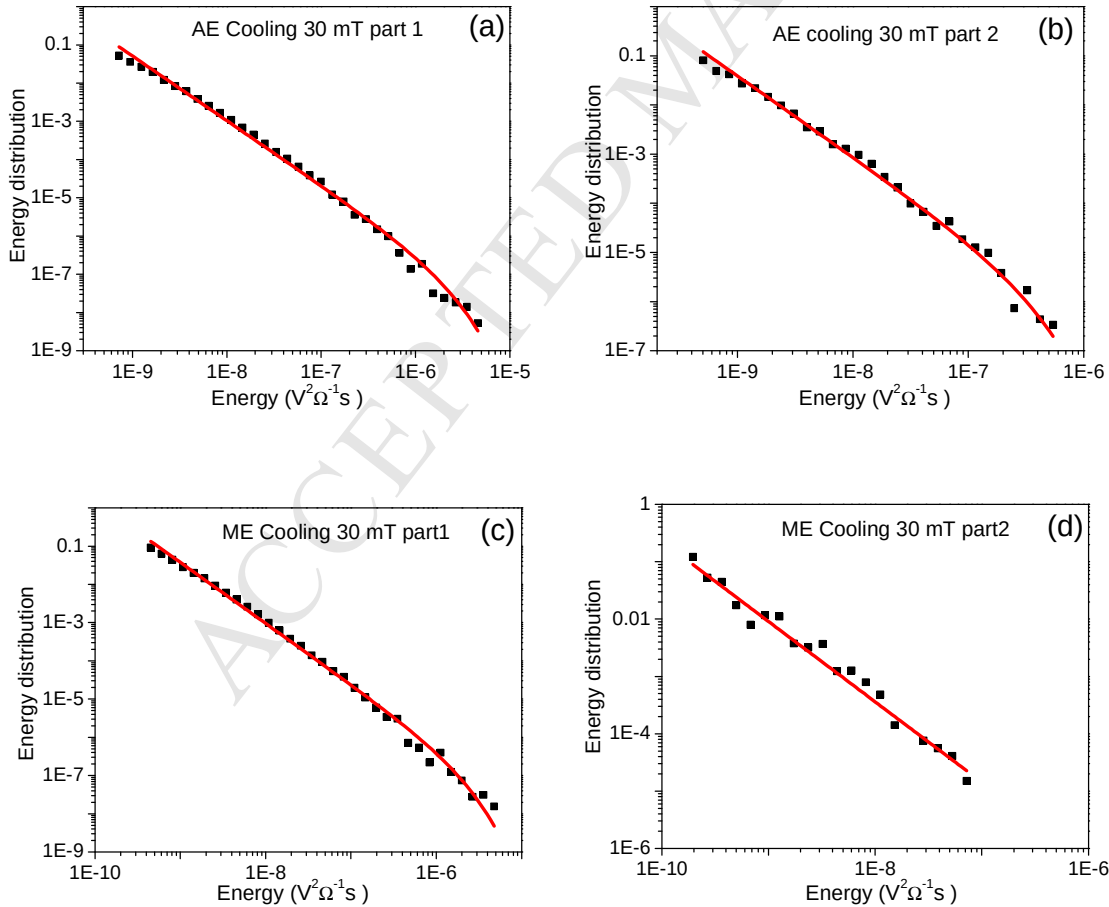


Fig. 14 Similar energy distribution functions as shown in Fig. 12 at $B=0$ mT; Log P versus log E, obtained from acoustic and magnetic measurements, during cooling in $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystals under the externally applied magnetic field ($B=30$ mT). The numbers of hits were 22187 for a), 2756 for b), 13683 for c) and 243 for d), respectively.

The energy exponents were the same in both parts for AE ($\varepsilon=1.7$ for the first part and $\varepsilon=1.7$ for the second part, $\Delta\varepsilon=0.1$) and differ only slightly from each other for ME ($\varepsilon=1.6$ for the first part and $\varepsilon=1.4$, for the second part, $\Delta\varepsilon=0.2$).

4. Discussion of the results

In metamagnetic SMAs, such as NiMnCoIn alloys studied here, forward and reverse martensitic transformations demonstrate, in general, different character: the forward transformation is usually isothermal while the reverse transformation displays an athermal character [40]. Such a response is usually not observed in conventional ferromagnetic SMAs. Therefore, it is critical to study the differences in the jerky transformation behaviour during both cooling and heating across the transformation and compare the exponents of power law distribution function capturing the jerky behaviour during both heating and cooling. This can help to better reveal the kinetics of the transformation and the nature of the avalanches. Table 3 displays the comparison of the energy exponents under zero external magnetic field obtained from thermal, acoustic and magnetic noises in three different ferromagnetic shape memory alloys investigated so far, including the present metamagnetic SMA.

Table 3. Energy exponents, ε , in three ferromagnetic shape memory alloys for heating (H) and cooling (C) cycles, obtained from thermal (DSC) acoustic emission (AE) and magnetic emission (ME) noises in Ni_2MnGa , $Ni_{49}Fe_{18}Ga_{27}Co_6$ and $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ single crystalline FMSMAs.

Ni_2MnGa [26,28] (austenite: cubic martensite: monoclinic)						$Ni_{49}Fe_{18}Ga_{27}Co_6$ [24,36,37] (austenite: $L2_1$, cubic martensite: $L1_0$, tetragonal, or 10M/14M modulated monoclinic + $L1_0$)						$Ni_{45}Co_5Mn_{36.6}In_{13.4}$ (austenite: $B2$ or $L2_1$, cubic martensite: $L1_0$, tetragonal, or 10M/14M modulated monoclinic or 10M/14M modulated monoclinic + $L1_0$)					
		AE		ME		DSC		AE		ME		DSC		AE		ME	
H	C	H	C	H	C	H	C	H	C	H	C	H	C	H	C	H	C
-	1.7	1.68	1.56	1.95	1.57	-	2.0	1.7	2.0	1.6	1.7	-	2.0	1.70	1.70	2.0	1.7
	$\pm$	$\pm$	$\pm$	$\pm$	$\pm$		$\pm$	$\pm$	$\pm$	$\pm$	$\pm$		$\pm$	± 0.1	± 0.1	$\pm$	$\pm$
	0.2	0.04	0.04	0.03	0.03		0.1	0.1	0.1	0.1	0.1		0.3			0.2	0.2

It can be seen that there is a positive asymmetry for Ni_2MnGa (the exponents are larger for heating than that for cooling), it is slightly negative for the $Ni_{49}Fe_{18}Ga_{27}Co_6$ and is practically zero for the present $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ metamagnetic SMA. Furthermore, the energy exponents, obtained from different noises, are almost the same within the error bars for the same direction in the three alloys shown in Table 3. While it is well established that the

energy exponents obtained from DSC and acoustic emissions are the same [25], the fact that the energy exponent of the magnetic noise is also the same can be an indication that the origin of all types of noises during martensitic transformation is the displacement discontinuities across the propagating interfaces.

Interestingly, there is no asymmetry in the energy exponents for the present metamagnetic SMA. Although the noise activities were larger for cooling, which would indicate a positive asymmetry (i.e. the exponents would be larger for heating than that for cooling [35]) this minor effect cannot be distinguished within the present error bars. It was observed in [40] that by interrupting the cooling (as also shown in Fig. 10), an increase of the amount of the transformed martensite fraction as a function of time was detected, which is typical for isothermal martensitic transformation. However, it was not clear in [40] whether the thermal activation assisted nucleation of the martensite and/or motion of the interphase boundaries was responsible for the isothermal increase of the martensite volume fraction. In the present experiments practically no AE and ME noise activities were detected during dwelling, while during further cooling clear acoustic and magnetic noises were obtained. The fact that the exponents were the same before and after dwelling indicates that before and after dwelling the same noisy growth process of the martensite takes place. This also means that the power law behaviour was confirmed in both parts and our energy distribution functions were stationary, similar to those cases where a lack of criticality was reported in other types of crackling noises (see e.g. [47]). The practically zero noise activity during dwelling in the present experiments can be rationalized only if our 5 min dwell time was not long enough for the transformation to continue (in [40] the martensite volume fraction grew from 8% up to 13% during about 30 min), or the further growth process during dwelling is different and much less noisy, or the cooling process is not isothermal. It is also worth noting that the asymmetry can also be different in polycrystalline and single crystalline samples (see e.g. [48]). Further investigations are needed to verify or negate the possibility of the above mechanisms.

Regarding the effect of the external magnetic field on the acoustic and magnetic noises, two major observations demand further discussion: i) increase in the magnetic noise activity under a magnetic field, and ii) the change in the polarity of the magnetic emission signals with increasing external magnetic field. In the present alloy, the austenite is ferromagnetic and the martensite is weakly magnetic. There is still a controversy concerning the magnetic state of the martensitic phase [11, 49, 50], however, the magnetic state of the weakly magnetic martensite plays a minor role in the magnetic noise, since all dominant changes of magnetization are related to change of the volume fraction of the strongly magnetic austenite. Indeed, the source of the magnetic emission can be correlated with the changes in magnetization as follows. In the absence of external magnetic field, the intermittent decrease of the fraction of the ferromagnetic austenite phase during cooling leads to magnetic signals in the detector coil in both directions (Fig. 15a), since the vanishing domains are randomly oriented and the magnetic domain distribution should correspond to an almost zero remnant magnetization (see also Fig. 1). Thus, in the detector coil bipolar signals should be detected, as observed here. In an external magnetic field of about 250 mT, the sample should be almost magnetically saturated and thus the abrupt jumps of the interface

should lead to the changes of magnetization in the same direction, i.e. unipolar signals should appear (Fig. 15b), which was the case as shown in Fig. 9. In addition, the amplitudes of the emission signals should be obviously larger in the presence of external magnetic field since the individual jumps of magnetization during a certain interface jump will be larger (the whole area of the flopping magnetization has the same orientation). Similar explanation holds for heating.

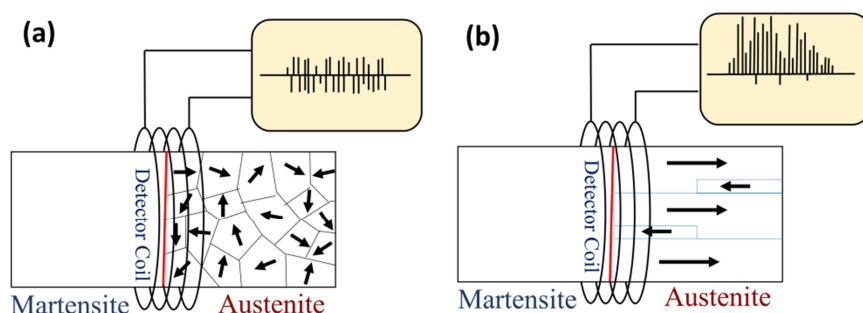


Fig. 15 Effect of external magnetic field on the polarity of the magnetic signals in the detector coil: the arrangement of magnetic domains in the austenite becomes more ordered with increasing field.

Summary and Conclusions

Characteristics of thermal, acoustic and magnetic noises, detected during martensitic transformation in the $Ni_{45}Co_5Mn_{36.6}In_{13.4}$ metamagnetic shape memory alloy single crystals, were investigated for the first time. The energy and amplitude distributions of the DSC and acoustic signals, as well as the energy, amplitude, area and the width (duration) distributions of the magnetic signals exhibit a power-law behavior.

The energy exponents obtained from the DSC measurements and acoustic emission signals are the same within the experimental errors ($\varepsilon=2.0\pm0.3$ and $\varepsilon=1.7\pm0.1$, respectively). The fact that the energy exponent of the magnetic noise is also the same can be an indication that the origin of all types of noises during martensitic transformation in these alloys is the displacement discontinuities across the propagating interface.

The exponents of the energy and amplitude distributions of the magnetic and acoustic signals were independent of the external magnetic field in the 0-230 mT range indicating that the character of the martensitic transformation is the same. Interestingly, however, these exponents during cooling and heating were the same in contrast to the expected asymmetry based the fact that the transformation has an isothermal character during cooling and an athermal one for heating [40]. Interrupting cooling and dwelling at a temperature above the martensite finish temperature, practically no acoustic and magnetic noises were detected, while with the continued cooling, the noise appeared again, with the same exponents. This, together with the fact the exponents were practically the same for heating and cooling, also indicates that the main discontinuous events, leading to noise emissions, should be the same in both directions.

Under zero external field, the magnetic noise packets contained voltage peaks in the detector coils in both directions. In small external magnetic fields (between 20 and 230 mT), the activity of magnetic signals increased and the magnetic peaks appeared more and more unidirectional (during both heating and cooling), which was rationalized taking into account the increasing magnetization of the austenite.

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Highlights

- Noisy character of martensitic transformation in a metamagnetic SMA
- Thermal, acoustic and magnetic noises were simultaneously measured
- Energy exponents are the same for all noises (belong to same discontinuous events)
- Exponents of magnetic and acoustic emission are independent of the magnetic field
- Magnetic noise becomes unipolar above 250 mT (magnetic saturation of the austenite)