ENGINEER

international scientific journal

ISSUE 3, 2026 Vol. 4
E-ISSN
3030-3893
ISSN
3060-5172



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ENGINEER

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E-ISSN: 3030-3893

ISSN: 3060-5172

VOLUME 4, ISSUE 3

SEPTEMBER, 2026

engineer.tstu.uz

TASHKENT STATE TRANSPORT UNIVERSITY

ENGINEER

INTERNATIONAL SCIENTIFIC JOURNAL
VOLUME 4, ISSUE 3 SEPTEMBER, 2026

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Tashkent State Transport University had the opportunity to publish the international scientific journal “Engineer” based on the **Certificate No. 1183** of the Information and Mass Communications Agency under the Administration of the President of the Republic of Uzbekistan. **E-ISSN: 3030-3893, ISSN: 3060-5172.** Articles in the journal are published in English language.

Journal Engineer ISSN: 3030-3893 Volume:4| Issue:3| 2026

Received: 30 July 2026 | **Accepted:** 10 September 2026 | **Published:** 30 September 2026

Structure, magnetostructural transformations and magnetocaloric properties of Heusler alloys and the MnAs compound: a review

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Abstract: The review systematises current views on the relationship between the crystal structure, the magnetic ordering and the magnetocaloric effect in two groups of promising functional materials — Heusler alloys of the Ni–Mn–Z (Z = Ga, In, Sn) families and the MnAs compound. Two principal approaches to the quantitative evaluation of the effect are discussed: the indirect one, based on the application of the Maxwell relations to magnetisation and heat-capacity data, and the direct one, which implies an immediate measurement of the adiabatic temperature change. It is shown that indirect methods are convenient for the screening of materials, but in the region of first-order phase transitions, where the conditions of applicability of the Maxwell relations are violated, the error of the calculated quantity may reach tens of per cent. The structural transformations in MnAs are considered separately — the first-order transition from the ferromagnetic hexagonal $B8_1$-type phase to the paramagnetic orthorhombic B31-type phase near 317 K and the subsequent second-order transition to the hexagonal paramagnetic phase near 394 K — together with the form of the magnetic phase diagram of this compound in steady fields. It is concluded that the combination of a high sensitivity of the magnetostructural transition to the external field with the proximity of the transition temperature to room temperature makes these materials the most realistic candidates for the working body of solid-state magnetic refrigerators.

Keywords: magnetocaloric effect, Heusler alloys, MnAs, magnetostructural phase transition, adiabatic temperature change, isothermal entropy change, Curie point, magnetic refrigeration, shape memory effect

1. Introduction

The development of functional materials whose physical properties change abruptly under a small external stimulus remains one of the actively developing directions of solid-state physics. Practically significant effects in such materials are, as a rule, realised near phase transitions, where the system becomes unstable with respect to weak perturbations. A characteristic example is the thermoelastic martensitic transformation observed in a number of intermetallic compounds, primarily nickel-containing ones: the rearrangement of the twin structure of martensite during such a transformation is accompanied by a considerable reversible change of the shape and dimensions of the specimen, which is known as the shape memory effect. Shape memory materials have found application in medical devices, instrument engineering and actuator systems.

Fundamentally new possibilities arise in those compounds in which the structural transformation occurs inside a magnetically ordered phase. In the Heusler alloy Ni_2MnGa the martensitic transition is realised in the ferromagnetic state, and as a consequence the lattice and the spin subsystems become rigidly coupled. Such a magnetostructural transformation can be induced not only by temperature but also by a magnetic field or by a mechanical stress, and it leads to a simultaneous change of the structure, the magnetisation, the electrical resistivity and the entropy of the specimen. It is with this strong spin–lattice coupling that the giant magnetoresistance, the magnetically driven shape memory effect [14] and the magnetocaloric effect observed in these materials are associated.

The present work is a critical review of published data on the structure, the character of the phase transformations and the magnetocaloric properties of Heusler alloys of the Ni–Mn–Z families and of the MnAs compound. The aim of the review is to compare the results obtained by direct and by indirect methods and to outline the range of problems that require further experimental study in high magnetic fields. The paper reports no new experimental data; its contribution consists in the systematisation of published results and in the identification of the quantitative discrepancy between the two measurement approaches, which is essential for the correct screening of candidate materials.

2. Research methodology

2.1. Scope of the review and selection of sources

The review covers publications devoted to three groups of questions: the thermodynamics and the measurement of the magnetocaloric effect; Ni–Mn–Z (Z = Ga, In, Sn) Heusler alloys together with their cobalt-doped quaternary modifications; and the MnAs compound with its substituted analogues. Preference was given to works that report either a direct measurement of the adiabatic temperature change or a joint analysis of magnetisation, heat-capacity and structural data, since only such works make a comparison of the two approaches possible. Lattice parameters and phase compositions are quoted as reported by the original authors. The quantities compared throughout the review are the adiabatic temperature change ΔT_{ad}, the isothermal change of the magnetic part of the entropy ΔS_m, the transition temperature and the width of the hysteresis loop.

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2.2. The magnetocaloric effect: definitions and thermodynamic background

The magnetocaloric effect consists in a change of the temperature of a magnetic material upon an adiabatic change of the external magnetic field or, under isothermal conditions, in the release or absorption of heat by the material. Accordingly, the effect is characterised by two interrelated quantities: the adiabatic temperature change ΔT_{ad} and the isothermal change of the magnetic part of the entropy ΔS_m. The sign of ΔT_{ad} allows the direct ($\Delta T_{ad} > 0$) and the inverse ($\Delta T_{ad} < 0$) magnetocaloric effect to be distinguished; the latter is typical of Heusler alloys with an inverse martensitic transformation, in which switching the field on stabilises the phase with the lower lattice entropy [12, 15].

The physical nature of the effect is related to the redistribution of entropy between the magnetic, the lattice and the electronic subsystems. Near the Curie point, that is, at a second-order phase transition, the field suppresses the fluctuations of the spin system and reduces the magnetic entropy; at a first-order transition the latent heat of the structural transformation is added to this, which is what provides the so-called giant magnetocaloric effect [4, 16].

2.3. Direct and indirect methods of evaluating the effect

Experimentally, the magnetocaloric effect is studied in two ways. The direct method implies the measurement of ΔT_{ad} by a thermocouple or a resistance thermometer attached to the specimen upon a rapid insertion of the specimen into the field or its rapid withdrawal. The indirect method is based on the calculation of ΔS_m from magnetisation isotherms by means of the Maxwell relation and, if heat-capacity data in a field are available, on the subsequent calculation of ΔT_{ad}. The indirect approach is less demanding with respect to instrumentation and therefore dominates in the literature, which makes it a convenient tool for the primary screening of materials [16, 17].

In the region of a first-order phase transition, however, where the magnetisation undergoes a discontinuity and the state of the specimen depends on its history, the applicability of the Maxwell relation is limited and the calculated values may be overestimated; according to published estimates, the discrepancy with direct measurements reaches 20–30 % [16, 17]. The spread of direct measurements is hindered by the absence of standardised procedures, by the difficulty of ensuring adiabatic conditions and by the limited availability of high fields: most works have been performed in fields of up to 2 T produced by permanent-magnet systems. For Heusler alloys and for MnAs, however, a field of 2 T is not sufficient for the complete completion of the first-order transition, and as a result the measured ΔT_{ad} turns out to be lower than the potentially attainable value. This circumstance determines the relevance of direct measurements in fields above 5–10 T.

3. Results and Discussion

3.1. Realisation of the effect in a refrigeration device

The practical realisation of the effect in a cooling device is as follows. The working body, being in a thermally insulated state, is introduced into the region of a strong field and is heated owing to the direct magnetocaloric effect; the excess heat is removed by a heat-transfer fluid to the hot heat exchanger. The working body is then withdrawn from the field, is demagnetised and cools below the initial temperature, after which it absorbs heat from the object to be cooled. Upon completion of the cycle the sequence is repeated, and the material operates as a magnetic heat pump. It is essential that a single change of the field produces a comparatively small temperature span; the widening of the working interval is achieved by regenerative schemes, in particular by the active magnetic regenerator (AMR) cycle, in which the temperature gradient is accumulated along the packed bed over many cycles [16, 17].

3.2. Heusler alloys of the Ni–Mn–Z families

Heusler alloys of composition Ni–Mn–Z (Z = Ga, In, Sn) and their quaternary modifications doped with cobalt are attractive because the temperature of the martensitic transition in them can be shifted towards room temperature by varying the composition, while the transition itself possesses a high sensitivity to the magnetic field. In the Ni–Mn–Sn and Ni–Mn–In–Co systems an inverse magnetocaloric effect is observed [12]; for the composition $Ni_{45.7}Mn_{36.6}In_{13.5}Co_{4.2}$ a direct measurement yielded $\Delta T_{ad} \approx -8$ K in a field of 1.95 T, which is one of the largest values of the inverse effect in moderate fields [13]. Alloys of the Ni–Mn–Ga family, on the contrary, exhibit predominantly a direct magnetocaloric effect, and it is in these alloys that the magnetically driven strain caused by the field-induced reorientation of martensite variants is realised [14].

The practical application of these materials is hindered by the hysteresis of the first-order transition, which lowers the reversibility of the effect upon cycling, and also by their brittleness and comparatively low thermal conductivity. Current research is therefore directed both at narrowing the hysteresis loop by means of alloying and heat treatment and at studying the irreversible component of the effect under repeated field cycling.

3.3. Structural and magnetic transformations in MnAs

The MnAs compound is a model object for the study of the spin–lattice interaction. At room temperature it is ferromagnetic and has a hexagonal structure of the $B8_1$ type (space group $P6_3/mmc$, α phase). On heating to T ≈ 317 K a first-order transition to the paramagnetic orthorhombic phase of the B31 type (space group Pnma, β phase) takes place, in which the magnetic ordering disappears simultaneously with the change of the lattice symmetry. The transformation is accompanied by a giant magnetostriction of the order of 2 % and by the release of a considerable latent heat. A further increase of the temperature to ≈ 394 K leads to a second-order displacive transition and to the restoration of the hexagonal symmetry, but now in the paramagnetic state (γ phase).

X-ray diffraction studies give for the hexagonal phase the lattice parameters $a \approx 3.730$ Å and $c \approx 5.700$ Å. In the region of the first-order transition temperature a coexistence of the phases is observed: according to quantitative phase analysis, at T = 317 K the fraction of the hexagonal phase is about 56 % and that of the orthorhombic phase about 44 %, which is typical of first-order transformations with a wide two-phase interval. The parameters of the orthorhombic phase are $a \approx 5.720$ Å, $b \approx 3.692$ Å, $c \approx 6.35$ Å.

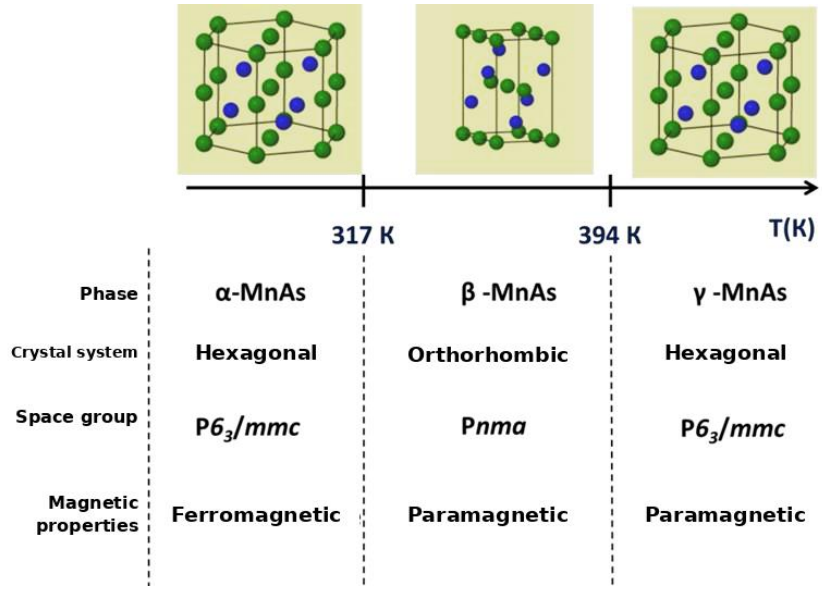


Fig. 1. Scheme of the structural and magnetic transformations in the MnAs compound

The theoretical description of the α → β transition has a long history. The first phenomenological model, proposed by Bean and Rodbell, started from an explicit dependence of the Curie temperature on the volume of the unit cell: for a sufficiently strong magnetoelastic coupling the transition changes from second order to first order with a jump of the volume [5]. Later an alternative mechanism was proposed which relates the transformation to a transition of the manganese ions from the high-spin to the low-spin state induced by a lattice distortion [6]. A subsequent treatment within the framework of the Landau theory with two coupled order parameters — the magnetisation and the lattice strain — made it possible to describe both transitions in a unified way and to reproduce the phase diagram with the lines of the first- and second-order transitions, including the influence of the field. The β → γ transition is thereby interpreted as a stabilisation of the hexagonal phase upon an increase of the volume caused by thermal expansion. Taken together, these results indicate the decisive role of lattice distortions and of the strong spin–lattice coupling in MnAs.

The magnetic phase diagram of the compound in a steady field has been constructed from magnetisation and volume-effect data. The region of existence of the orthorhombic paramagnetic phase is bounded in the (T, μ₀H) plane by fields of approximately up to 9 T: the line of the first-order transition, which starts from the point T ≈ 317 K at H = 0, and the line of the second-order transition, which starts from the point T ≈ 394 K, converge at a tricritical point near 370 K. The presence of a tricritical point means that the very character of the transformation can be controlled by an external field, converting it from first order into second order — a circumstance of fundamental importance for reducing the hysteresis in practical applications.

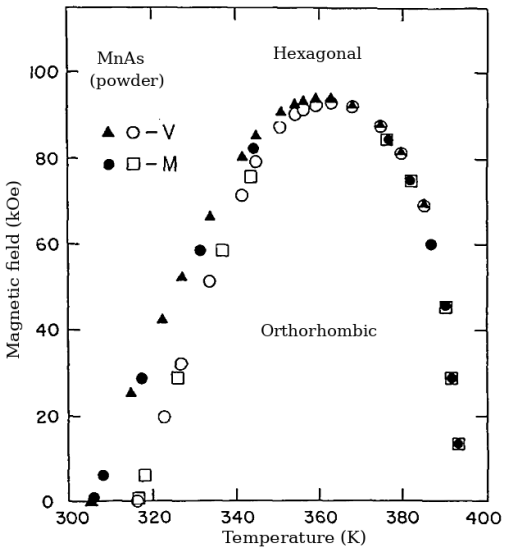


Fig. 2. Magnetic phase diagram of MnAs in a steady magnetic field, constructed from the magnetisation M and the volume V of the crystal lattice. Filled symbols denote the transition from the orthorhombic to the hexagonal structure, open symbols the reverse transition

https://doi.org/10.56143/3030-3893-2026-3-163-167

3.4. Development of magnetocaloric research and the current state of the problem

The effect was discovered by E. Warburg in 1881 [1]. The practical use of magnetic cooling was first demonstrated in 1933 by Giauque and MacDougall, who by the method of adiabatic demagnetisation of gadolinium sulfate reached a temperature of about 0.25 K, a record low value for that time [2]. The possibility of operation near room temperature was shown by Brown in 1976 [3]: using metallic gadolinium and up to fifty successive magnetisation–demagnetisation cycles in fields of up to 7 T, he obtained a temperature span between the hot and the cold ends of the device of about 47 K, that is, approximately three times larger than the ΔT_{ad} of gadolinium for a single change of a field of the same magnitude.

A qualitatively new stage began in 1997 with the discovery of the giant magnetocaloric effect in the compound $Gd_5Si_2Ge_2$, caused by a first-order magnetostructural transition [4]. Subsequently the giant effect was found in $La(Fe,Si)_{13}$ compounds and their hydrides [10, 11], in $MnFeP_{1-x}As_x$ compounds [9], in MnAs and its substituted analogues [7, 8], in Heusler alloys of various families [12, 13] and in $MnCoGeB_x$ systems. The effect is also studied intensively in manganites, where it serves as a sensitive probe of the critical behaviour near the magnetic ordering temperature [18].

The interest in this subject over the last three decades is due to two circumstances. From the fundamental point of view the magnetocaloric effect is a sensitive tool for the investigation of magnetic and magnetostructural transformations, since it is directly related to the entropy of the system. From the applied point of view, a significant share of the world consumption of electric energy falls on the production of artificial cold and on air conditioning, and the transition to solid-state magnetic cooling technologies that do not use gaseous refrigerants with a high greenhouse potential could yield a gain both in energy efficiency and in environmental safety [16, 17].

3.5. Comparative assessment and open questions

The comparison of the two groups of materials shows that they are limited by different factors. In Ni–Mn–Z Heusler alloys the transition temperature is controlled over a wide range by the composition, but the reversibility of the effect is restricted by the hysteresis and by the brittleness of the intermetallic compound. In MnAs the transition lies close to room temperature by its very nature and the field sensitivity of the transformation is high, but the working point can be shifted only by substitution or by external pressure. Common to both groups is the principal methodological problem: the values of ΔS_m obtained from the Maxwell relation cannot be regarded as a reliable characteristic of a first-order transition, and the published values of ΔT_{ad} have been measured, for the most part, in fields that are insufficient to complete the transition. Consequently, a direct comparison of materials from data of different origin is at present of limited validity, and the cyclic rather than the single-shot value of the effect should be adopted as the criterion of practical suitability.

4. Conclusion

1. The analysis of published data shows that the most realistic candidates for the role of the working body of a room-temperature magnetic refrigerator remain materials with a first-order magnetostructural transition near 300 K that combine a high sensitivity of the transition temperature to the field with a moderate hysteresis. These requirements are met to the greatest extent by Heusler alloys of the Ni–Mn–Z families and by the MnAs compound.

2. The indirect method based on the Maxwell relation is convenient for the primary screening of materials, but in the region of a first-order transition it may overestimate the effect by 20–30 %; the values obtained in this way should therefore be verified by direct measurements.

3. Most of the published direct measurements have been performed in fields of up to 2 T, which is insufficient for the complete completion of the transition in the materials considered; the accumulation of direct ΔT_{ad} data in fields above 5–10 T is a priority experimental task.

4. Since the suitability of a material for a device is determined by the cyclic and not by the single-shot value of the effect, the study of the reversibility of the effect under repeated field cycling should be regarded as an equally necessary part of the characterisation of a magnetocaloric material.

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