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Heat capacity, thermal conductivity and magnetocaloric effect in Heusler alloy $Ni_{47}Mn_{40}Sn_{13}$

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Received September 28, 2022
Revised September 28, 2022
Accepted September 29, 2022

The results of a study of heat capacity, thermal conductivity and magnetocaloric effect of the polycrystalline $Ni_{47}Mn_{40}Sn_{13}$ alloy depending on temperature ($T = 80-350$ K) and magnetic field ($0-8$ T) are given. A considerable difference between the values of a sudden change in heat capacity ΔC_P in the heating and cooling mode was found near the magnetostructural martensite–austenite phase transition (MSPT), which is related to influence of latent heat of phase transition. Thermal conductivity in the range of $T = 80-300$ K rises with temperature ($d\kappa/dT > 0$) and increases in more than three times. Electronic thermal conductivity in the martensitic phase ($T = 150$ K) is 37% of the total value. An anomalous rise of thermal conductivity $\Delta\kappa = \kappa(\text{aust}) - \kappa(\text{mart}) = 4.2$ Wm K was found in the region of MSPT. The contributions of electrons and phonons to the observed sudden change are 63 and 37% respectively and are conditioned both by a rise of mobility of conduction electrons under a martensite–austenite transition and by an increase of phonons' free path length.

The magnetocaloric effect in cyclic magnetic fields with the amplitude of 1.8 T was studied. A dependence of reverse effect value on temperature scanning rate was established. The direct measurements of ΔT in the cyclic magnetic field of 1.2 T show a twofold decrease of the effect amplitude near T_C at an increase of cyclic magnetic field frequency from 1 to 30 Hz. Most likely, this is related to magnetic and microstructural heterogeneities which act as an additional thermal dissipation channel.

Keywords: Heusler alloys, heat capacity, thermal conductivity, magnetocaloric effect, cyclic magnetic fields.

DOI: 10.21883/PSS.2022.12.54407.487

1. Introduction

The sustained interest in studies of Heusler alloys of the Ni-Mn-X (X = Ga, Sn, In) family is directly related to their unique physical properties, which are most pronounced in the region of phase transformations under the influence of external impacts (magnetic field, temperatures, pressure etc). A sequence of magnetic, structural, electronic change of phase transitions, which sometimes occur simultaneously and cause the so-called magnetostructural analysis phase transition (MSPT), is observed in Heusler alloys. This unique phenomenon is due to a strong interrelation of magnetic and lattice subsystems of a solid body. In this case, the alloy structure and magnetic properties change under the impact of a magnetic field, and the following effects are observed: magnetoresistance, a giant value of the magnetocaloric effect, magnetically controlled effect of shape memory. All this arouses keen interest of researchers.

An essential factor, stimulating the theoretical and experimental studies of magnetostructural phase transitions and associated phenomena, is the possibility of practical application of materials with MSPT in modern technologies, in particular, as a working medium in the magnetic cooling technology. The operating efficiency of a cooling machine based on the magnetocaloric effect (MCE) is determined, among other things, by the rate of heat withdrawal from the cooling machine working medium. Obviously, the operating efficiency of a cooling machine depends not only on MCE value and cyclic frequency of applied magnetic field, but also on value of thermal conductivity of the cooler working medium material: the higher its thermal conductivity, the higher the cooling machine efficiency. Hence, a conclusion can be made that, from the viewpoint of applied problems, the need for studying thermal conductivity of promising functional materials is undisputed. Moreover, measurement of thermal conductivity is very nearly the only method for determining the acting mechanisms of dissipation of heat carriers in a solid body, and this is a fundamental problem of condensed matter physics.

Phase transitions, magnetic and structural properties in the studied Ni-Mn-Sn system have been studied theoretically and experimentally in sufficient detail [1–10]. Papers [11–15] are dedicated to a detailed study of the structure, magnetic and magnetocaloric properties of the studied compound $Ni_{47}Mn_{40}Sn_{13}$. A magnetic paramagnetic-ferromagnetic transition is first observed (at a temperature decrease) in the given composition at $T_C \approx 313\,K$, which takes place in the austenitic phase. A further temperature decrease is accompanied by a magnetostructural transition from the highly-symmetrical cubic austenite (ferromagnetic) to low-temperature monoclinic or orthorhombic martensite ($M_S \approx 200\,K$), while magnetic state of martensite is heterogeneous.

Significantly less attention is paid to thermophysical properties of Heusler alloys than to other physical properties. We know only of several papers which give the results of thermal conductivity study [16–22]. These papers are characterized by a relatively low thermal conductivity value, comparable values of phononic and electronic components and various anomalies observed near the temperatures of magnetic and structural phase transitions.

The authors of [16] studied thermal and electrical conductivity of several samples of the Ni-Mn-In alloy in the range of $T = 130-530\,K$. They have found a sharp rise of thermal conductivity near the martensitic transition and associate the revealed phenomenon with an increase of electronic thermal conductivity under a martensite–austenite transition thanks to an increased mobility of charge carriers. A similar conclusion for the $Ni_{48}Mn_{39}Sn_{13}$ sample is given in [23].

The authors of [18] explain the sudden rise of thermal conductivity of the $Ni_{2+x}Mn_{1-x}Ga$ alloy near the MSPT temperature by changes in the alloy's phononic subsystem. According to [18], the phonon spectrum degrades near the MSPT and additional phononic excitations arise; they correspond to the acoustic branch of phonons and lead to a rise of heat capacity C_P and, consequently, κ_{ph}. The sharp thermal conductivity peak $\frac{\Delta\kappa}{\kappa} \approx 70\%$ near T_C in the $Ni_{50}Mn_{34}In_{16}$ alloy found in [20] is explained by the authors by changes in the electronic spectrum of current carriers under a paramagnetic–ferromagnetic phase transition. The authors of [22] associate the anomalous behavior of thermal conductivity and temperature of conductivity near the martensite–austenite phase transition in $Ni_{50}Mn_{28}Ga_{22-x}(Cu,Zn)_x$ alloys with coexistence and competition of phases. The aforesaid results of the studied of $\kappa(T)$ for several Heusler alloys indicate the absence of a common understanding of peculiarities of the heat transfer mechanism in alloys and need for continuation of such studies.

The present paper gives the results of an experimental study of thermophysical (with a detailed analysis of mechanisms of heat carrier scattering and influence of a magnetic field on them) and magnetocaloric properties of Heusler alloy $Ni_{47}Mn_{40}Sn_{13}$. The direct studies of MCE were performed both by the extraction method in stationary magnetic fields up to 1.8 T, and by the method of magnetic field modulation in iterative magnetic fields of 1.2 and 1.8 T.

2. Sample and measurement procedure

A sample was obtained by the mechanical fusing method [12]. Thermal conductivity was measured by the continuous heat flux method. Heat capacity — by the A.c.-calorimetry method. Magnetocaloric effect — by the magnetic field modulation method. Chromel–constantan and copper–constantan thermocouples Ø = 0.0 5mm were used as temperature sensors. The sample is a rectangular plate sized $\approx 7 \times 3 \times 1\,mm^3$. In order to exclude the influence of hysteresis effects, the sample was cooled down to liquid nitrogen temperatures prior to measurements in the heating mode, and in case of cooling — to temperatures above T_C.

3. Results and discussion

Temperature dependence of heat capacity in the heating and cooling mode is shown in Fig. 1. Two anomalies near the structural ($T_S = 220\,K$) and magnetic ($T_C = 313\,K$) phase transitions and a hysteresis near T_S are seen. Moreover, the anomaly at T_S almost disappears under at reverse variation of temperature. The crystal lattice abruptly expands when heated near T_S, this requires additional energy equivalent to latent heat of a phase transition, and that's why C_P rises. The cooling process, on the contrary, is associated with energy release, that's why the amount of outside heat, required for changing the sample temperature, decreases, while ΔC_P decreases. A magnetic field shifts the structural transition temperature T_S towards low temperatures, suppresses fluctuations of the magnetic order parameter near T_C and smooths the peak in dependence $C_P(T)$ near T_C. A similar pattern for the $Ni_{50}Mn_{37}(In_{0.2}Sn_{0.8})_{13}$ alloy was observed by the authors of [10] who also relate the observed phenomenon with influence of latent heat of a phase transition.

Let us consider the behavior of the temperature dependence of thermal conductivity of the $Ni_{47}Mn_{40}Sn_{13}$ alloy. Thermal conductivity of magnetic materials in the general case is a sum: $\kappa_{tot} = \kappa_e + \kappa_{ph} + \kappa_m$, where κ_e, κ_{ph} and κ_m

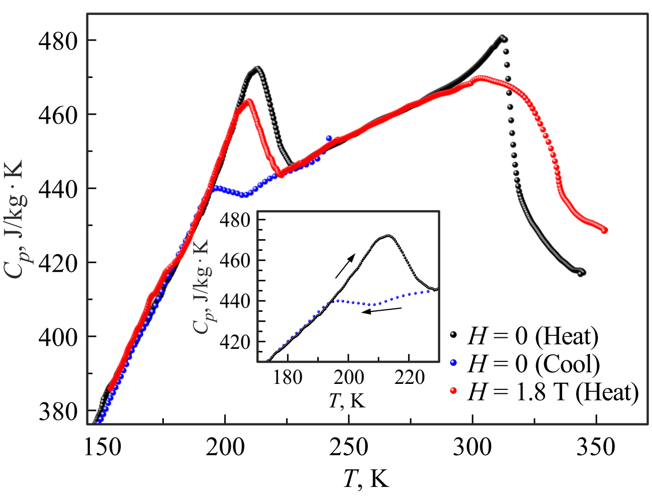

Figure 1. Temperature dependence of heat capacity in the heating and cooling mode. The red dots show the data in the magnetic field of 1.8 T in the heating mode.

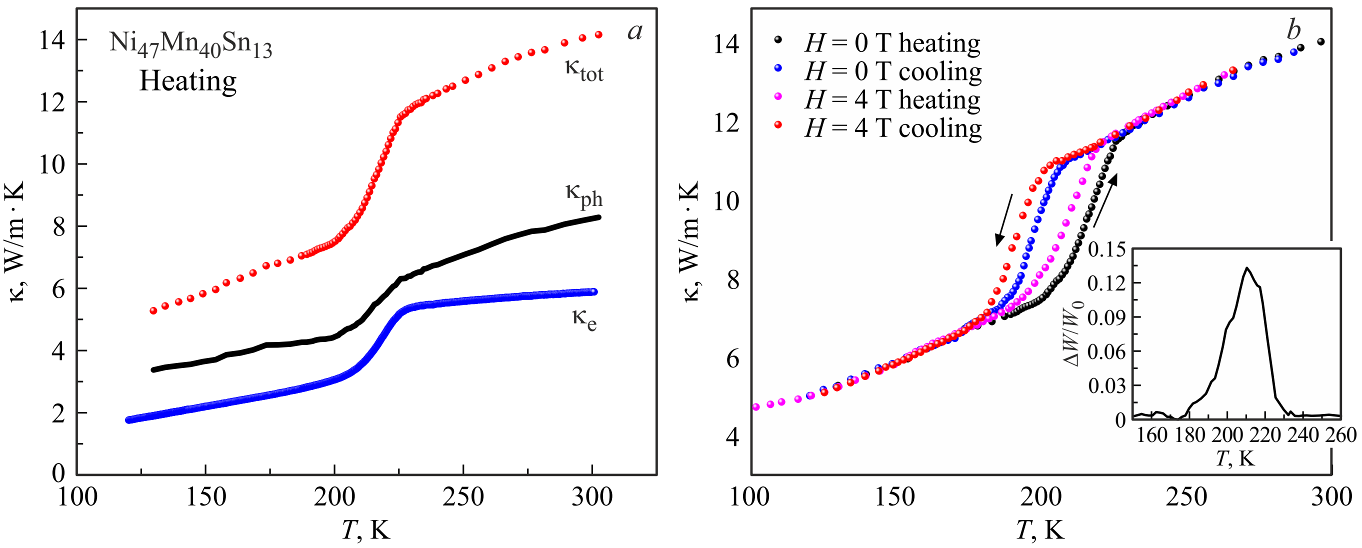

Figure 2. *a* — temperature dependence of the common, phononic and electronic components of thermal conductivity; *b* is influence of magnetic field on thermal conductivity. The inset shows thermal magnetoresistance.

are the electronic, phononic and magnonic components of thermal conductivity respectively. The magnetic component is usually ignored when analyzing $\kappa_{tot}(T)$ due to its small contribution to thermal conductivity [24]. The electronic component is determined from the Wiedemann–Franz ratio $\kappa_e = LT/\rho$, assuming that $L = L_0 = 2.24 \cdot 10^{-8}\,V^2/K^2$ is the Lorentz number. This assumption is righteous for high-resistive metal alloys [25]. Fig. 2, *a* shows the temperature dependence of the common, phononic and electronic components of thermal conductivity. Phononic contribution κ_{ph} was obtained as the different between the measured value of κ_{tot} and electronic κ_e calculated from the Wiedemann–Franz ratio. The electrons' contribution to the common thermal conductivity is 37% (at $T = 150$ K).

Attention should be paid to the following peculiarities in the behavior of κ_{tot}. Firstly, this is linear growth of thermal conductivity with temperature $d\kappa/dT > 0$ in the wide temperature range of 100–300 K, which is not typical for crystalline solid bodies, where phonon-phononic changeover processes start prevailing in this region. The second peculiarity is an anomalously abrupt increase of thermal conductivity in the region of a martensite–austenite phase transition. In principle, the observed sudden rise of κ_{tot} can be related both to an increase of κ_e under a transition and to a rise of κ_{ph}. An anomalous rise of thermal conductivity from 7 to 11.5 W/m K under a martensite–austenite transition was observed by the authors of [16] in the Ni-Mn-In alloy. Such behavior is mainly associated with a rise of the electronic component thanks to a rise of electron mobility upon sample's transition to a more ordered phase. A similar sudden change of thermal conductivity $\Delta\kappa_{tot} = 2$ W/m K for the $Ni_{1.92}Mn_{1.56}Sn_{0.52}$ alloy was found by the authors of [23] who also ascribe the observed effect to the electronic contribution. This explanation in our case is not completely correct, because a sudden change $\Delta\kappa_e = \kappa_e(\text{aust}) - \kappa_e(\text{mart}) = 2.6$ W/m K, while the total change of thermal conductivity at MSPT is $\Delta\kappa_{tot} = \kappa_{tot}(\text{aust}) - \kappa_{tot}(\text{mart}) = 4.2$ W/m K. This means that a phononic contribution to the sudden change is significant and equal to $\Delta\kappa_{ph} = 1.6$ W/m K and they cannot neglected.

The following can be stated as regards the reasons for a linear growth of $\kappa_{ph}(T)$. It is known that phonon-phononic scattering processes (changeover processes) prevail in solid bodies near and above the Debye temperature and $\kappa_{ph} \sim T^{-1}$. The absence of this kind of dependence $\kappa_{ph}(T)$ means that other mechanisms can prevail in the course of phonon scattering, in particular, phonon scattering on small-scale structural imperfections of the crystal lattice. For such dissipation to occur, the structural heterogeneities where phonons scatter must be comparable to the average free path length of phonons. Let us estimate the free path length of phonons, using an expression for phononic thermal conductivity: $\kappa_{ph} = \frac{1}{3} C_P l_{ph} v_S d$, where v_S — sound velocity, d — sample density, κ_{ph} — experimental value of thermal conductivity, C_P — specific heat capacity. Using the experimental values of C_P and κ_{ph} (Fig. 1 and 2) and literature data $v_S = 4500$ m/s [26], $d = 8.2\,g/cm^3$ [21], we find $l_{ph}(\text{mart}) = 8.3$ Å, $l_{ph}(\text{aust}) = 12.4$ Å, i.e. there is an abrupt, almost by 50%, increase of the phonons' free path length, thus causing the observed sudden change $\kappa_{ph}(T)$ near the MSPT. Local distortions of the crystal lattice and nanosized twin boundaries, concentration of which rises upon a transition to the martensitic phase [16,27], can be considered as a mechanism leading to a sudden change of l_{ph}.

It should be also noted that phonons effectively disperse on obstacles, size of which are comparable to the wavelength of thermal phonons which corresponds to the given temperature. Wavelength λ_{ph} can be estimated using

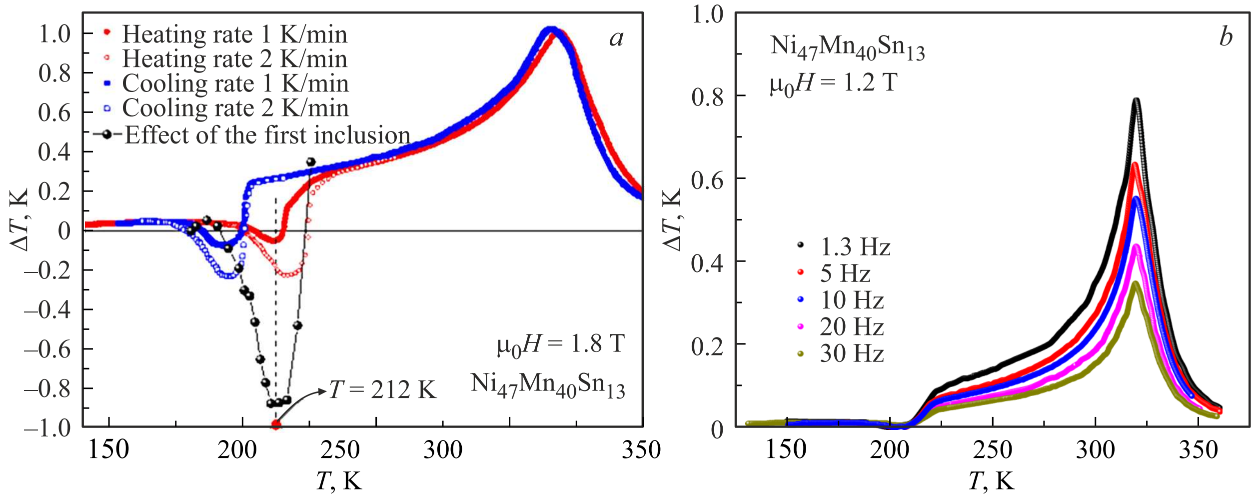

Figure 3. *a* — temperature dependence of MCE in the heating and cooling mode in the alternating magnetic field of 1.8 T at $f = 0.2$ Hz. The data of the direct MCE measurements upon single activation of the magnetic field of 1.8 T are also given there for comparison (black dots). *b* is the frequency dependence for $Ni_{47}Mn_{40}Sn_{13}$ in the cyclic field of 1.2 T in the range of 1−30 Hz.

the formula $\lambda_{ph} = 2a\,\frac{\theta}{T_S}$, where a is the lattice constant, T_S is the MSPT temperature. The estimations show that $\lambda_{ph} \approx 18$ Å, which is comparable to l_{ph}. In order to explain the observed course of $(d\kappa/dT) > 0$, it must be assumed that l_{ph} is a quantity independent from T, which is possible when l_{ph} is limited by sizes of crystal lattice structural imperfections, with which phonons interact, and v_S — is slightly dependent on temperature. It is seen that the values of l_{ph} are comparable to sizes of structural units of the crystal lattice and suddenly increase upon a transition to a more ordered and symmetrical phase. The temperature course of thermal conductivity in this case is determined by the dependence of heat capacity on temperature, which is seen in Fig. 1 and 2 where $C_P \sim T$, $\kappa_{tot} \sim T$, $\kappa_{ph} \sim T$ between phase transition temperatures.

A distinctive feature of the behavior of thermal conductivity of materials with MSPT is the dependence of the phononic component on magnetic field. The volume ratio of austenite with a highly-symmetrical cubic structure increases under the influence of a magnetic field near the MSPT. The rate of electron and phonon scattering decreases accordingly, which leads to a rise of κ_{tot}. Fig. 2, *b* shows the dependence of $\kappa_{tot}(T)$ in the magnetic field of 4 T in the heating and cooling mode. A magnetic field leads to a shift of the phase transition temperature towards low temperatures and a rise of thermal conductivity. Similarly to the magnetoresistive effect, the thermal magnetoresistive effect can be estimated

$$\frac{\Delta w}{w_0} = \frac{w_H - w_0}{w_0} = \frac{\kappa_0 - \kappa_H}{\kappa_H}, \qquad (1)$$

where w is the thermal resistance ($w = 1/\kappa$). As seen from the figure (inset), the value of the thermal magnetoresistive effect in the field of 4 T reaches $\approx -13\%$.

Fig. 3, *a*−*b* shows the results of the direct studies of MCE of the $Ni_{47}Mn_{40}Sn_{13}$ alloy in cyclic magnetic fields. The magnetocaloric properties of the $Ni_{47}Mn_{40}Sn_{13}$ sample in pulsed and stationary magnetic fields were studied in [11–13]. Fig. 3, *a* shows the temperature dependence of MCE at different rates of sample heating/cooling $V = 1$ and 2 K/min in cyclic magnetic fields with the amplitude of 1.8 T and frequency of $f = 0.2$ Hz. As seen in Fig. 3, the dependence of $\Delta T(T)$ features a direct MCE ($\Delta T > 0$) at T_C and reverse MCE ($\Delta T < 0$) under a magnetostructural phase transition. A negative sign of MCE means that antiferromagnetic correlations are present in the martensitic phase. An increase of the temperature scanning rate leads to an increase of the reverse effect value. The nature of such behavior is considered in more detail in [6,28,29]. The maximum reverse effect value obtained by the extraction method (upon single activation of a magnetic field) in the magnetic field of 1.8 T is equal to −0.9 K (see the points in Fig. 2). It is seen that the maximum value of the reverse effect during measurement by the modulation method is considerably smaller than in case of a single magnetic field activation. This is due to the fact that the effects of the first activation are not measured during measurements in cyclic fields, a signal is generated on the basis of several cycles. Therefore, the slower the change of sample temperature in the region of a magnetostructural analysis phase transition, the more field activation/deactivation cycles take place and, accordingly, the greater part of the sample passes into the austenitic phase, and the observed reverse effect value is small. It means that a significant value of MCE under cyclic application of a magnetic field cannot be obtained in the studied material in the hysteresis region, due to an irreversible martensite–austenite transition. Fields which shift the transition temperature beyond the temperature hysteresis are required to obtain a reversible MCE. It has been demonstrated in [6] that the magnetic field value of 8 T for a similar alloy $Ni_{47}Mn_{40}Sn_{12.5}Cu_{0.5}$ is small for inducing a reversible MSPT.

Fig. 3, *b* shows the results of the measurement of influence of the cyclic magnetic field frequency on the MCE value in the field of 1.2 T. It is seen that the effect value near T_C decreases from 0.78 to 0.35 K, i.e. more than twofold, when frequency increases from 1 to 30 Hz. The reverse effect is almost absent near the MSPT at these frequencies (there is only a break in the MCE temperature dependence) due to the above-mentioned reason. It is known that values of adiabatic temperature change near second-order phase transitions must not depend on cyclic magnetic field frequency. Indeed, on the one hand, the recent studies of dependence $\Delta T(T, f)$ for Gd in the magnetic field of 1.2 T showed a frequency stability of MCE value in the interval of $1-20$ Hz [30]. On the other hand, paper [31] notes a slight decrease of amplitude ΔT_{ad} for Gd with an increase of the magnetic field modulation amplitude from 116 to 1184 Hz in the field of 33 mT. The frequency studies of MCE in various magnetocaloric materials show a complex pattern of MCE dependence on cyclic field frequency, even for materials with second-order phase transitions [30–33]. While the dependence of ΔT on magnetic field frequency near first-order phase transitions can be explained by growth and nucleation (phase transition kinetics), the situation is not so evident for second-order phase transitions. A decrease of amplitude ΔT_{ad} with a rise of cyclic magnetic field frequency is not an artefact, but is a result of interaction between several temperature-dependent magnetic and thermal properties and requires further studying. Most likely, the degree of effect amplitude decrease will also depend on magnetic field frequency and intensity.

4. Conclusion

The results of the studies of thermophysical and magnetocaloric properties of the $Ni_{47}Mn_{40}Sn_{13}$ alloy can be summarized as follows. The differences in values of a sudden change of heat capacity ΔC_P in the heating and cooling mode $\Delta C_{P\,\text{heat}} > \Delta C_{P\,\text{cool}}$, found in dependence $C_P(T)$, are related to the influence of latent heat of a phase transition. The temperature coefficient of thermal conductivity in the region of studied temperatures $T = 80-350$ K has a positive sign $(d\kappa/dT > 0)$, while the electronic and phononic fractions of thermal conductivity are $\approx 37\%$ and 63% respectively. An abrupt rise of thermal conductivity $\Delta\kappa = \kappa(\text{aust}) - \kappa(\text{mart}) = 4.2$ W/m K was found near the MSPT temperature. The contributions of electrons and phonons to the observed sudden change are 63 and 37% respectively and are conditioned both by a rise of mobility of conduction electrons and by an increase of phonons' free path length as a result of the martensite–austenite transition. The thermal magnetoresistive effect $\frac{\Delta W}{W_0} = \frac{\kappa_H - \kappa_0}{\kappa_H}$ was studied. It was shown that the effect value reaches 13% in the field of 4 T.

The magnetocaloric effect in cyclic magnetic fields with the amplitude of 1.8 T was studied. It was shown that the reverse effect value depends on heating/cooling rate. With a rise of frequency of the cyclic 1.2 T magnetic field, quantity ΔT near T_C decreases approximately twofold when cyclic magnetic field frequency increases from 1 to 30 Hz. This fact must be studied further in greater detail and might be related to magnetic and microstructural heterogeneities which act as an additional thermal dissipation channel.

Funding

The study was funded by a grant of the Russian Science Foundation No. 22-19-00610.

Conflict of interest

The authors declare that they have no conflict of interest.

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