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MAGNETISM

Experimental Simulation of a Magnetic Refrigeration Cycle in High Magnetic Fields

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Abstract—The complete magnetic refrigeration cycle has been simulated on a sample of gadolinium in magnetic fields of a Bitter coil magnet up to 12 T. The total change of temperature of the sample during the cycle is a consequence of magnetic refrigeration, and the dependence of the magnetization of the sample on the magnetic field exhibits a hysteretic behavior. This makes it possible to determine the work done by the magnetic field on the sample during the magnetic refrigeration cycle and to calculate the coefficient of performance of the process. In a magnetic field of 2 T near the Curie temperature of gadolinium, the coefficient of performance of the magnetic refrigeration is found to be 92. With an increase in the magnetic field, the coefficient of performance of the process decreases sharply down to 15 in a magnetic field of 12 T. The reasons, for which the coefficient of performance of the magnetic refrigeration is significantly below the fundamental limitations imposed by the reversed Carnot theorem, have been discussed.

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1. INTRODUCTION

The magnetocaloric effect consists in changing the temperature of a magnetic material when it is magnetized or demagnetized in an external magnetic field. The magnetocaloric effect is characterized by a change of the temperature under adiabatic conditions (ΔT) or by a quantity of heat (ΔQ) absorbed or released by a magnetic material under isothermal conditions with a change of the magnetic field. The highest values of the magnetocaloric effect are achieved near the points of phase transitions in magnetic materials. In this connection, at present a large number of new magnetic materials with related phase transitions of different nature have been synthesized and investigated [1–6]. Great hopes are placed on the use of materials with the magnetocaloric effect as a basis of the new technology of solid-state heat engines, such as refrigerators and heat pumps operating near room temperature, which are alternative to traditional freon-based engines [7–10].

The most important characteristic of refrigerating machines is the coefficient of performance of the process (COP) of magnetic refrigeration. This coefficient is calculated as the ratio of the quantity of heat transferred from a cold reservoir to the amount of work expended during one thermodynamic cycle (Fig. 1). The coefficient of efficiency of the refrigerating machine with a working body based on a magnetic material is calculated according to the formula

$$\varepsilon = \frac{Q_2}{\delta A}, \quad (1)$$

where Q_2 is the quantity of heat transferred from the cold reservoir during the cycle and δA is the work done by the magnetic field on the magnetic material during the cycle. The work is defined in terms of the external magnetic field H and the magnetization M as follows:

$$\delta A = \mu_0 \oint H dM. \quad (2)$$

In order to achieve the maximum coefficient of efficiency of the refrigeration system, it is necessary that the coefficient of performance of the process should tend to values corresponding to the ideal refrigeration cycle, i.e., to the coefficient of performance of the process of the ideal reversed Carnot cycle, which is calculated according to the formula

$$\varepsilon_0 = \frac{T_2}{T_1 - T_2}, \quad (3)$$

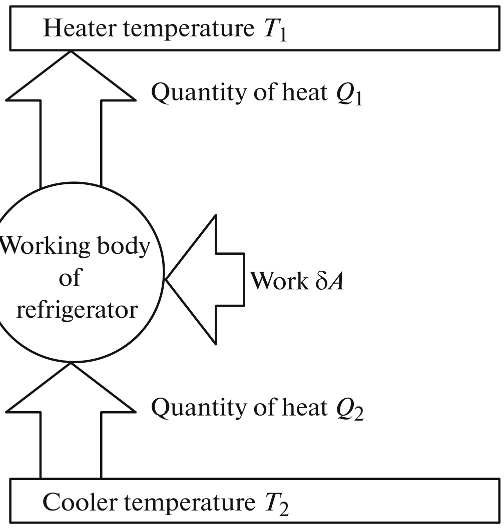

Fig. 1. Principle of operation of the refrigerating machine based on the reversed Carnot cycle. The quantity of heat Q_1 is transferred by the working body to the heater, the quantity of heat Q_2 is supplied to the working body from of the refrigerator, and the amount of work δA is expended during the cycle.

where T_1 and T_2 are the operating temperatures of the hot and cold reservoirs, respectively. The ratio of the two coefficients of performance determines the perfection factor of the thermodynamic cycle:

$$\eta = \frac{\varepsilon}{\varepsilon_0}100\%. \quad (4)$$

2. MATERIALS AND EXPERIMENTAL TECHNIQUE

For the purpose of experimentally studying the magnetic refrigeration and measuring the coefficient of performance of the process, we designed and constructed the experimental setup for the simultaneous measurements of the magnetocaloric effect and magnetization of the samples of magnetic materials as a function of the magnetic field. In addition, we developed a special insert into the Bitter coil magnet, which made it possible to simultaneously measure the magnetocaloric effect of two samples of magnetic materials, one of which was under quasi-adiabatic conditions (ΔT), whereas the other sample was under quasi-isothermal conditions (ΔQ), as well as to measure the magnetization of the former sample as a function of the magnetic field. The measurements were performed in the magnetic fields ranging from 2 to 12 T.

Figure 2 shows a three-dimensional model of the insert into a Bitter coil magnet with the samples under investigation. The insert is placed inside the vacuum chamber in the magnetic field of the Bitter coil magnet. The characteristic time of thermal relaxation of the samples inside the vacuum chamber is approximately 300 s. A thermal sensor glued to sample *1* is a platinum resistance thermometer measuring a change of the temperature of the sample ΔT. Simultaneously, the magnetization of sample *1* is measured using two Hall sensors located near the "equator" (sensor *5*) and the "pole" (sensor *6*) of the sample in the direction perpendicular to the magnetic force lines (Fig. 3). The difference between the readings of the two Hall sensors is the useful signal, which is proportional to the magnetization of the sample in the magnetic field. After the additional calibration against the isothermal magnetization measured by the standard method (Fig. 4), the useful signal makes it possible to calculate the work done by the magnetic field on the material according to formula (2) from the magnetization/demagnetization of the sample during one on/off cycle of the magnetic field.

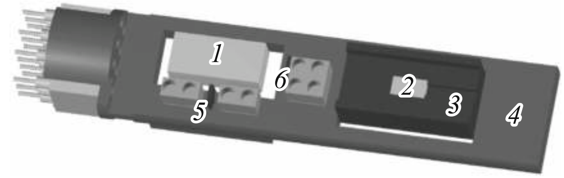

Fig. 2. A three-dimensional model of the Bitter coil magnet insert: (*1*) sample of the material with the magnetocaloric effect under the quasi-adiabatic conditions, (*2*) sample of the material with the magnetocaloric effect under the quasi-isothermal conditions, (*3*) massive nonmagnetic substrate with a good thermal conductivity, (*4*) plastic frame holder, (*5*) "equatorial" Hall sensor, and (*6*) "pole" Hall sensor.

Pure gadolinium is chosen as the material for the experimental simulation of the magnetic refrigeration. Gadolinium is one of the most thoroughly investigated magnetocaloric materials in the world [11–13]. As is known, this material exhibits a maximum direct magnetocaloric effect near the Curie point $T_C = 294$ K. In

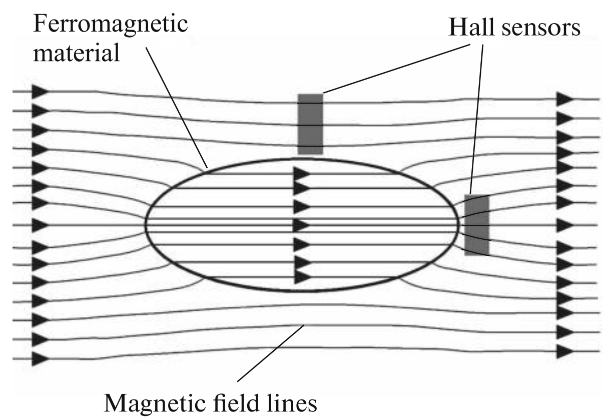

Fig. 3. Physical principle of the measurement of the magnetization of a ferromagnet in a magnetic field with the use of two Hall sensors.

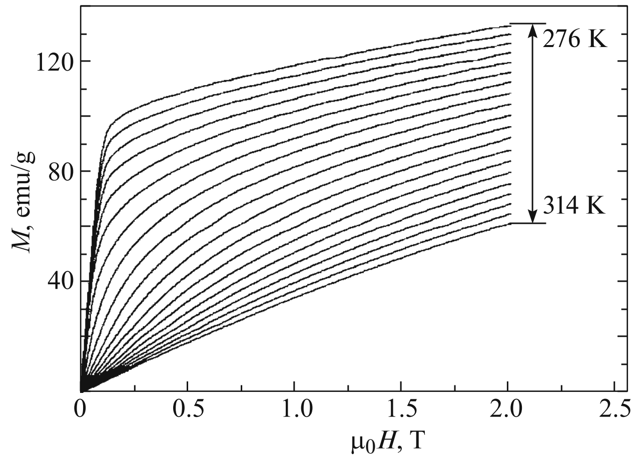

Fig. 4. Field dependences of the magnetization of gadolinium measured at different temperatures in the range from 276 to 314 K with a step of 2 K under the isothermal conditions.

this study, we used the gadolinium sample with a weight $m = 3.572$ g.

Sample *2* was in good thermal contact with massive nonmagnetic substrate *3*, i.e., under the quasi-isothermal conditions. During the experiment, we measured the change in the substrate temperature and calculated the isothermal heat flux ΔQ, i.e., the magnetocaloric effect under the isothermal conditions. Details of the experiments on measurements of the magnetocaloric effect in different materials according to the above technique were described in [14–16]. The obtained data on the investigation of the magnetocaloric effect in gadolinium were presented in [17].

For the thermodynamic system used in this work, the limiting coefficient of performance of the Carnot process can be calculated using formula (3), where T_1 is the temperature maintained in the vacuum chamber (the initial temperature of the sample) and T_2 is the temperature of the sample at the end of the refrigeration cycle (at the moment when the magnetic field is decreased to 0 T). Let the change in the temperature of the sample during the cycle be $\Delta T_{\text{cycle}} = T_2 - T_1$. Then, knowing the specific heat c of the sample and the sample weight m, the quantity of heat transferred from the cold reservoir (sample) during the cycle can be expressed as

$$Q_2 = mc\Delta T_{\text{cycle}}. \quad (5)$$

Of course, in formula (5), we did not take into account the dependence of the specific heat of the sample on the temperature and magnetic field, but, for gadolinium, the error due to the change in the specific heat in the magnetic field does not exceed 10% [11].

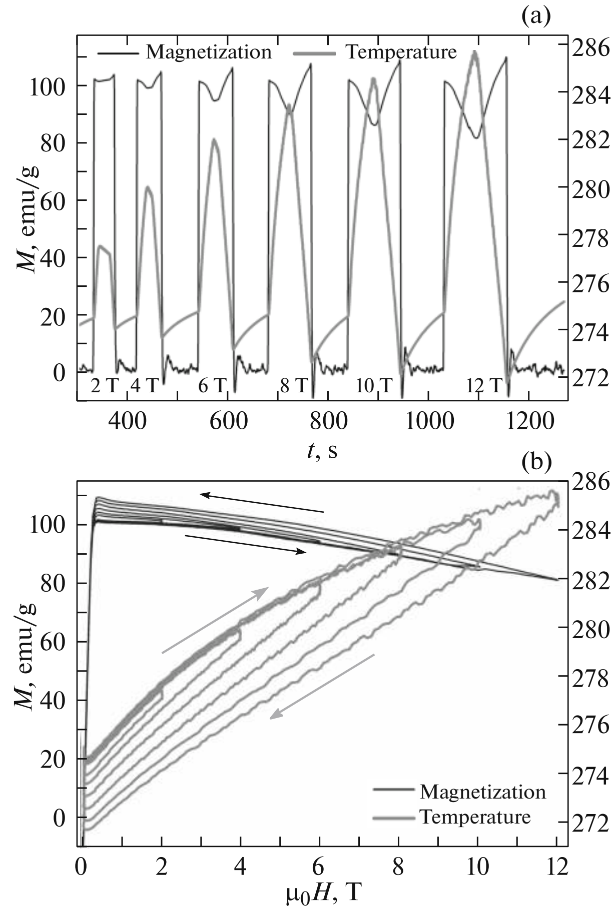

Fig. 5. (a) Time dependences of the magnetization and temperature of the sample in different magnetic fields and (b) dependences of the magnetization and temperature of the sample on the magnetic field under the quasi-adiabatic conditions. The initial temperature is 275 K.

3. EXPERIMENTAL RESULTS

The experiments were performed at two different initial temperatures of 275 and 298 K. Let us consider one on/off cycle of the magnetic field. It can be seen from Fig. 5a ($T_1 = 275$ K) that, with an increase in the magnetic field, the temperature of the sample increases, i.e., there is a direct magnetocaloric effect, which is accompanied by an increase in the magnetization of the sample; however, as the Curie temperature is approached, the magnetization begins to increase. The subsequent decrease in the magnetic field leads to a decrease in the temperature of the sample below the initial temperature T_1. This is explained by an enhanced heat transfer from the sample in a heated state into the external environment, whereas the magnetization exhibits an abrupt increase in magnitude with the subsequent decrease.

The results of the experiments at $T_1 = 275$ K (Fig. 5a) demonstrated that the temperature of the sample increases from 275 to 286 K ($\Delta T = 11$ K) with

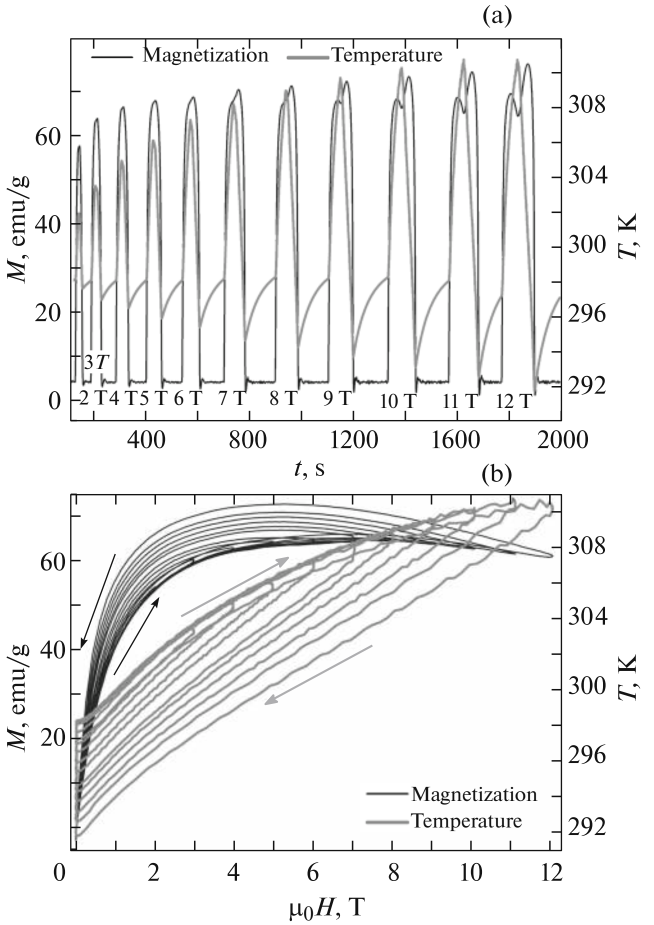

Fig. 6. Same as in Fig. 5, but at the initial temperature of 298 K.

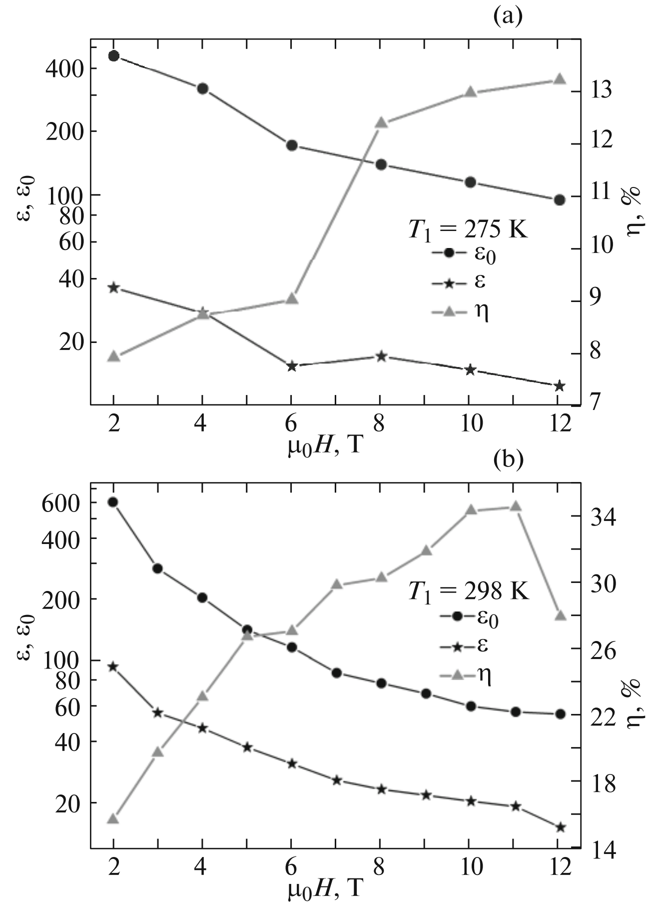

Fig. 7. Coefficients of performance of magnetic refrigeration in the real cycle (ε) and the ideal reversed Carnot cycle (ε_0) on a logarithmic scale and the perfection factor of magnetic refrigeration (η) as a function of the magnetic field at the initial temperatures of (a) 275 and (b) 298 K.

an increase in the magnetic field to 12 T and decreases from 286 to 272 K as the magnetic field decreases to 0 T; i.e., the resulting change in the temperature of the sample during the cycle $\Delta T_{\text{cycle}} = -3$ K is a consequence of the magnetic refrigeration. For the initial temperature $T_1 = 298$ K (Fig. 6a) in the same field of 12 T, the resulting change in the temperature of the sample is $\Delta T_{\text{cycle}} = -5.5$ K, which makes it possible to calculate the quantity of heat transferred from the sample during the cycle according to formula (5). It should be noted that, in both cases, the dependence of the magnetization of the sample on the magnetic field exhibits a hysteretic behavior (Figs. 5b, 6b). This is explained by a strong heating of the sample with the magnetocaloric effect and by a high rate of heat transfer into the external environment, which, after turning off the magnetic field, leads to a magnetic refrigeration of the sample. By calculating the integral of *HdM* over the closed contour, we can determine the work done by the magnetic field on the material during the cycle according to formula (2) and to calculate the coefficient of performance of the magnetic refrigeration according to formula (1).

For the initial temperature $T_1 = 275$ K, we obtained the coefficients of performance of the magnetic refrigeration ε = 36 in the magnetic field $\mu_0 H = 2$ T and ε = 12 in the magnetic field $\mu_0 H = 12$ T (Fig. 7a). For the initial temperature $T_1 = 298$ K, which is close to the Curie temperature T_C of gadolinium, the coefficient of performance of the magnetic refrigeration is found to be ε = 92 in the magnetic field $\mu_0 H = 2$ T and, with an increase in the magnetic field, decreases sharply to the value of ε = 15 in the magnetic field $\mu_0 H = 12$ T (Fig. 7b). The coefficient of performance of the magnetic refrigeration in magnetic fields of 2–12 T as a function of the amplitude of the magnetic field at different initial temperatures of 275 and 298 K is plotted on the logarithmic scale in Figs. 7a and 7b, respectively. It can be seen from these figures that, in both cases, the real coefficient of performance of the process has an exponential dependence on the magnetic field. A similar dependence qualitatively follows from the reversed Carnot theorem, which imposes the fundamental limitations on the coefficient of performance of any cyclic process of cooling. In particular, at the initial temperature $T_1 = 298$ K in the magnetic field $\mu_0 H = 10$ T, the real coefficient of performance of the process is equal to 20, whereas the coefficient of performance of the Carnot process is 59; accordingly, the perfection factor of the refrigeration cycle in the case under consideration is $\eta = 34\%$ (Fig. 7b).

The experimental values of the coefficient of performance of the process and the perfection factor indicate that the achievable values of the coefficient of performance of the process for magnetic refrigerators based on gadolinium in a magnetic field of 10 T are no less than 15–20. By varying the parameters of the real refrigeration cycle, or, more precisely, by bringing them closer to the parameters of the reversed Carnot cycle [18], it is possible to significantly improve the perfection factor ($\eta \sim 100\%$), for example, by means of the very slow turning-on of the magnetic field (isotherm) and the subsequent sharp turning-off of the field (adiabat).

4. CONCLUSIONS

The setup described in this paper provides a means for simultaneously measuring the magnetocaloric effect and magnetization of a sample of a magnetic material as a function of the magnetic field of a Bitter coil magnet in the range up to 12 T under the quasi-adiabatic conditions. It was shown that the quasi-adiabatic change in the magnetization of a gadolinium sample in a magnetic field exhibits a hysteretic behavior. This is explained by the magnetocaloric effect and the heat transfer into the external environment, which results in a magnetic refrigeration of the sample. According to the obtained data, the coefficients of performance of the magnetic refrigeration in a magnetic field of 10 T at the initial gadolinium sample temperatures of 275 and 298 K are equal to 15 and 20, respectively. From the comparison of the coefficients of performance of the real magnetic refrigeration cycle and the ideal reversed Carnot cycle, it was found that the perfection factors of the magnetic refrigeration at temperatures of 275 and 298 K in a magnetic field of 10 T are equal to 13% and 34%, respectively. In order to increase the perfection factor of the magnetic refrigeration, it is necessary that the experimental conditions should be as much as possible close to the parameters of the reversed Carnot cycle.

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