A Setup for Direct Measurement of the Adiabatic Temperature Change in Magnetocaloric Materials

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Abstract—In order to find a highly efficient, environmentally-friendly magnetic refrigerant, direct measurements of the adiabatic temperature change \( \Delta T_{\text{adb}} \) are required. Here, in this work, a simple setup for the \( \Delta T_{\text{adb}} \) measurement is presented. Using a permanent magnet Halbach array with a maximum magnetic field of 1.8 T and a rate of magnetic field change of 5 T/s, an accurate determination of \( \Delta T_{\text{adb}} \) is possible in this system. The operating temperature range of the system is from 100 to 400 K, designed for the characterization of materials with potential for room temperature magnetic refrigeration applications. Using the setup, \( \Delta T_{\text{adb}} \) of a first-order and two-second-order compounds have been studied. Results from the direct measurement for the first-order compound have been compared with \( \Delta T_{\text{adb}} \) calculated from the temperature and magnetic field-dependent specific heat data. By comparing results from direct and indirect measurements, it is concluded that for a reliable characterization of the magnetocaloric effect (MCE), direct measurement of \( \Delta T_{\text{adb}} \) should be adopted.

Index Terms—Direct measurement of adiabatic temperature change, first-order phase transition, indirect measurement of adiabatic temperature change, magnetocaloric effect (MCE), second-order phase transition, specific heat, uncertainty in adiabatic temperature measurement.

I. INTRODUCTION

SINCE 1997, after the observation of the giant magnetocaloric effect (MCE) near room temperature in Gd\(_3\)(Si\(_2\)Ge\(_2\)) [1], several thousands of research papers have been published in search of a suitable magnetic refrigerant which can be applied in room temperature magnetic refrigerators. Unfortunately, till today no material has been found that is both commercially viable and environmentally friendly. The MCE is an intrinsic property of a magnetic material originating from the spin-phonon interaction of the material. In the case of adiabatic conditions the total entropy of a system, being the sum of its phonon and spin (or magnetic) entropies, is conserved. Therefore, changing the magnetic entropy by the application or removal of a magnetic field will change the phonon entropy and temperature of the system. This change in temperature is known as the adiabatic temperature change \( \Delta T_{\text{adb}} \) and the measurement of \( \Delta T_{\text{adb}} \) is known as the direct measurement of the MCE. Isothermal magnetization measurements yielding information about the isothermal entropy change \( \Delta S_M \) upon application or removal of a magnetic field can also be used to estimate the MCE indirectly. In addition, using data from temperature and magnetic field-dependent specific heat \( (C_H) \) measurements, the values of both \( \Delta S_M \) and \( \Delta T_{\text{adb}} \) can be calculated. The measurement of \( \Delta S_M \) and \( C_H \) is known as the indirect measurement of the MCE. Owing to the widespread availability of systems used for magnetization and specific heat measurements often indirect measurements of the MCE have been reported. A keyword search on 19th October 2022, in the “Web of Science” yields around 8929 publications where the keyword “magnetocaloric” is mentioned and among them only around 179 publications mention “magnetocaloric” and “direct” “adiabatic temperature.” These numbers are approximate but give evidence of that reports of direct measurements of \( \Delta T_{\text{adb}} \) are scarce. Moreover, several publications [2], [3], [4], [5], [6] report that \( \Delta T_{\text{adb}} \) estimated from indirect measurements differs largely from that of direct measurements owing to nonadiabatic conditions and approximations involved in indirect measurements. Pecharsky and Gschneider Jr. [7] have estimated the uncertainty involved in the indirect measurement process and found that it can be as large as \( \sim 15\% \) for elemental Gd. Furthermore, the values are highly sensitive to any approximation used in the calculation. By comparing direct and indirect MCE measurements, Pecharsky and Gschneidner [8] have also concluded that, in the indirect measurement, there will be around \( 1\% \sim 1.5\% \) uncertainty in the determination of \( \Delta T_{\text{adb}} \) near room temperature. It can therefore be argued that reliable estimation of \( \Delta T_{\text{adb}} \) requires direct measurements.

A few other techniques have been reported for the direct measurement of \( \Delta T_{\text{adb}} \). Infrared characterization techniques mostly applied at or close to room temperature have been demonstrated [9], [10], [11], [12]. Although the infrared technique is advantageous in terms of being a noncontact measurement, the comparatively narrow temperature range for reported systems poses a limitation. There are also commercial systems available based on a permanent magnet as a magnetic field source and thermocouples as temperature sensors, e.g., the AMT&C LLC system [13]. However, a detailed description of the different parts of the system, the heat transfer procedure, and a comparison between different measurement protocols are missing.

Here, in this work, we demonstrate a simple setup for the direct measurement of \( \Delta T_{\text{adb}} \). Using this setup, we have...
compared the direct and indirect measurements of the MCE of the first-order material La$_{0.7}$Ca$_{0.3}$MnO$_3$ (LCMO). Also, we have reported the direct MCE results for the second-order material La$_{0.8}$Sr$_{0.2}$MnO$_3$ (LSMO). In addition, we have measured $\Delta T_{\text{adb}}$ of a gadolinium sample and compared it with previously reported results.

II. INSTRUMENTATION

A $\Delta T_{\text{adb}}$ measurement process consists of three important steps; first adiabatic conditions should be established, second the magnetic field should be changed and measured, and third the temporal variation of the sample temperature should be monitored. These three steps have been incorporated into our setup. To create adiabatic conditions for the measurements, a high vacuum chamber ($\sim 10^{-6}$ hPa) is used. Along with the high vacuum chamber, the sample is wrapped with a layer ($\sim 2$ mm) of Pyrogel$^\text{1}$ [see Fig. 1(a)] to reduce the heat transfer rate from the sample, which provide sufficient time for the measurement of $\Delta T_{\text{adb}}$ upon a change of the magnetic field. A schematic of the sample rod is shown in Fig. 1(a). The sample rod is placed inside the vacuum chamber, which is kept inside a liquid N$_2$ filled cryostat. Therefore, the system can operate at temperatures above the boiling point of liquid N$_2$.

The magnetic field variation was controlled by a Halbach-type permanent magnet array [14], [15], which can produce a maximum magnetic field of 1.8 T. To measure the magnetic field accurately at the position of the sample, a calibrated Hall sensor is mounted on the sample rod, in a way that the magnetic field is perpendicular to the Hall sensor [see Fig. 1(a)]. Although the heat radiation and convection are much reduced, the heat conduction from the sample cannot be completely eliminated. This limits the overall measurement time. The measurement time is highly influenced by the magnetic field sweep rate, i.e., if the magnetic field sweep rate is low compared to the heat conduction rate from the sample, some amount of heat will be lost during the measurement. If the heat loss of the system is denoted by $x$, the measured adiabatic temperature change $\Delta T_{\text{measured}}$ for a magnetic field change of $\Delta H$ and a magnetic field sweep rate of $y$ can be

\[^{1}\text{Registered trademark.}\]
expressed as

$$\Delta T_{\text{measured}} = \Delta T_{\text{adb}} - \frac{\Delta H}{y} \times x. \quad (1)$$

Therefore, a large value of $y$ ensures a smaller difference between the actual and measured value of $\Delta T_{\text{adb}}$.

Khovaylo et al. [2] showed that a minimum magnetic field sweep rate of about 3 T/s was required for a correct determination of $\Delta T_{\text{adb}}$ in the system designed by them. Using our system, the temporal variations of the sample temperature, $\Delta T_S(t)$ have been measured for different magnetic field sweep rates for a first-order material, LCMO. The measurements were performed at a temperature near the magnetic ordering temperature $T_C$ (251 K) of the material with the highest available magnetic field change (1.79 T). The recorded data are presented in Fig. 1(b). To describe the effect of magnetic field sweep rate, the temporal variation of the sample temperature has been normalized with the value of $\Delta T_{\text{adb}} [2.12(\pm 0.01) \text{ K}]$ defined as the largest change of the sample temperature induced by a magnetic field change. From Fig. 1(b), it is clear that a minimum magnetic field sweep rate of about 4 T/s is required to neglect the heat loss during the measurement process. However, this value is not universal and it will vary with the heat loss rate of the system, the heat capacity of the sample, the response time of the temperature sensor, etc. [16]. Noticeably, a higher magnetic field sweep rate allows a longer time period for the $\Delta T_{\text{adb}}$ measurement. Therefore, all results presented in the following discussion have been obtained using a magnetic field sweep rate of 5 T/s. This rate is high enough to neglect the heat dissipation during the magnetization or demagnetization process.

The temperature of the sample is monitored and controlled by a commercially available temperature controller (LakeShore 335). As temperature sensors, T-type thermocouples (accuracy of $\sim 0.01 \text{ K}$) are used. As mentioned earlier, the response time of the temperature sensor plays a crucial role, the sensor system used here has a response time of less than 200 ms. A resistive Manganin heater providing a maximum power of 50 W is used as a heat source. Both in thermocouples and in heater, twisted types of wires have been used in order to reduce any noise produced by the induced stray magnetic field in the wires [17]. To understand the heat transfer process in the system, a block diagram and its equivalent electrical circuit are presented in Fig. 1(c) and (d), respectively. The sample with heat capacity $C_S$ and temperature $T_S$ is attached to a thermocouple via a thermal link (Ag-paint) with a thermal conductivity of $K_{\text{st}}$ being proportional to the contact area. Therefore, the temperature of the thermocouple $T_t$ differs slightly from the sample temperature $T_S$ and this difference depends upon the heat capacity of the thermocouple $C_t$. In the equivalent circuit, the inverse of the thermal conductivity represents a resistance, the heat capacity corresponds to a capacitance, and the flow of heat is described by electrical current. Thus, the heat flow across the sample due to the change of magnetic field has been replaced by a current source. Moreover, there is heat flow from the sample to the liquid $N_2$ chamber through the Pyrogel (with thermal conductivity $K_P$), and this heat flow is being controlled by the resistive heater; all these heat flows are collectively replaced by another current source in the equivalent electric circuit diagram. When the sample is subjected to a magnetic field change, either applied or removed, the temperature change of the sample is represented as $\Delta T_{\text{adb}}$, i.e., the real value of the adiabatic temperature change, while the measured temperature change across the thermocouple is represented as $\Delta T_{\text{adb}}^m$. These two quantities correspond to voltages across the capacitances $C_S$ and $C_t$ in the equivalent circuit and are related by the following equation:

$$\Delta T_{\text{adb}}^m = \Delta T_{\text{adb}} \frac{C_S}{C_S + C_t}. \quad (2)$$

Therefore, for an ideal measurement of $\Delta T_{\text{adb}}$, the value of $C_S$ should be much larger than the value of $C_t$. One way to satisfy this condition is to use a larger mass of the sample compared to the mass of the thermocouple. Porcari et al. [18] has demonstrated experimentally the effect of sample mass on the $\Delta T_{\text{adb}}$ measurement. As our thermocouple has a mass of about 0.1 mg and the dimensions of the sample space are $\approx 4 \times 1 \times 0.5 \text{ cm}^3$, a wide range of sample mass can be measured with good accuracy. Apart from the sample mass, the thermal conductivity of the thermal link (Ag-paint) plays a crucial role in determining the time response of the $\Delta T_{\text{adb}}$ measurement. The time ($t$) response from the RC-circuit can be expressed as

$$\Delta T_{\text{adb}}^m = \Delta T_{\text{adb}}^0 (1 - e^{-t/\tau}) \quad (3)$$

where $\tau$ is the time constant of the thermocouple. $\tau$ determines the rate at which thermocouple temperature $T_t$ will reach the sample temperature $T_S$. From the equivalent RC-circuit, $\tau$ can be expressed as

$$\tau = \frac{C_t}{K_{\text{st}}}. \quad (4)$$

Cugini et al. [16] have pointed out that $K_{\text{st}}$ is proportional to the sample-to-sensor contact area, thus a smaller contact area will increase the value of $\tau$. Near room temperature, considering a T-type thermocouple and the silver paint as a thermal link, the value of $\tau$ is $< 10^{-3} \text{ s}$. From Fig. 1(b) it is clear that the value of $\Delta T_{\text{adb}}^m$ is stable over a time period of a few seconds, which proves that our measurement process is reliable.

Although Pyrogel is required to establish adiabatic conditions, it creates a time lag between the thermocouple attached to the sample and the heat source (resistive heater) which affects the process of temperature control. To overcome this problem, a second thermocouple has been introduced outside the Pyrogel attached to the brass sample holder. For further discussion, the thermocouple attached to the sample is named as thermocouple B, while the thermocouple attached to the sample holder will be referred to as thermocouple A. To stabilize the sample temperature at any desired temperature and in order to not overshoot the desired temperature, first, the system temperature as measured by thermocouple A is stabilized at a temperature sweep rate of $\approx 1 \text{ K/min}$, followed by a waiting time of a $\sim 10 \text{ min}$ until thermocouple B also shows the desired temperature and has been stable at this temperature for a few minutes.
For the measurements in the direct measurement setup, two compounds have been selected, LCMO, which shows a first-order magnetic phase transition from the paramagnetic (PM) to ferromagnetic (FM) state at a temperature around 250 K, and LSMO, which shows a second-order PM to FM phase transition at a temperature around 325 K. In addition, measurements have also been performed on a 99.95% pure gadolinium sample. We have chosen two oxide materials instead of any intermetallic compound, as the oxide materials are advantageous owing to their chemical stability, ease of synthesis, resistance to corrosion, etc. Also, the magnetocaloric intermetallic compounds are highly sensitive to the synthesis process and often incorporate more than one impurity phase. For instance, the giant magnetocaloric compound FeMnP exhibits a $T_C$ of $\sim 390$ K when synthesized by the drop synthesis method [19], [20], while $T_C$ decreases to $\sim 300$ K when synthesized using ball-milling [21]. In addition, first-order intermetallic compounds with strong structural transition can suffer from a “virgin” effect, which can cause cracks on the sample surface and it will be difficult to maintain proper thermal contact between the sample and the temperature sensor [22]. Both oxide materials studied in this work were synthesized using a modified sol-gel method described in detail elsewhere [23]. Before describing the measurement results of these compounds, a description of the $\Delta T_{adb}$ measurement process will be presented.

Two types of measurement protocols have been used in this study, a loop measurement protocol and a phase reset protocol. The loop measurement protocol [24] corresponds to a fast measurement process of $\Delta T_{adb}$, which involves a continuous magnetization and demagnetization process while cooling or heating the compound. A schematic of the measurement protocol is presented in Fig. 2(a) for the cooling cycle of the LCMO compound. Before starting the loop measurement, the sample temperature was stabilized at $T_1$ in zero applied magnetic field. At $T_1$, a positive magnetic field ($\mu_0 H_f$) was applied raising the temperature of the sample to $T_1 + \Delta T_1$; this is indicated as step 1 in Fig. 2(a). In the second step, the magnetic field was changed to zero and consequently the sample temperature decreased from $T_1 + \Delta T_1$ to $T_1$. Steps 3 and 4 correspond to repetitions of steps 1 and 2, but this time a negative magnetic field with the same magnitude ($-\mu_0 H_f$) was applied and subsequently removed. The temperature sensor has a response time of about 200 ms and with a magnetic field sweep rate of 5 T/s it takes around 360 ms to achieve 1.8 T. In addition, the thermal conductivity of the sample can introduce an additional delay in the measurement. Therefore, although the measurement starts collecting data from 300 ms after the application/removal of a magnetic field, with a time interval between data points of 250 ms, we only consider data measured between 1000 and 3000 ms in the analysis. In addition, in order to verify that there is no background contribution disturbing the measurement a simultaneous measurement with thermocouple A (the chamber temperature) is also performed. Fig. 2(b) shows the final values of $\Delta T_{adb}$ at 251 K for the LCMO compound with cyclic field changes from 0 to 1.79 T (step 1), 1.79 to 0 T (step 2), 0 to $-1.79$ T (step 3), and finally $-1.79$ to 0 T. The second measurement protocol, the phase reset protocol, is a modified version of the loop measurement protocol. In the phase reset protocol, the process for the application and removal of the magnetic field is the same as previously described for the loop measurement protocol. However, before stabilizing the sample at the temperature $T_1$, the sample is heated (cooled) to a temperature in the PM (FM) state for cooling (heating) cycle measurements. Carron et al. [25] have shown that the field and temperature hysteresis of a first-order compound plays an important role and without adopting the phase reset protocol, the calculation of the isothermal entropy change using Maxwell’s relation often generates a large uncertainty. The uncertainty involved in the direct measurement of $\Delta T_{adb}$ is related to the accuracy of the absolute temperature measured by the thermocouple, the magnetic field dependence of the thermocouple, and possible heat dissipation during measurement due to any nonadiabatic condition. However, as the thermocouple mass is negligible compared with the mass of the sample and a T-type thermocouple with negligible magnetic field dependence is used, the uncertainty is significantly reduced. The negligible effect of magnetic field on the thermocouple has been verified by performing temperature-dependent loop measurements without

![Fig. 2. (a) Real-time loop measurement protocol of magnetic field dependent temperature change for the LCMO compound. (b) Magnetic field dependent average values of $\Delta T_{adb}$ for different directions of the magnetic field change. Arrows indicate the magnetic field change direction.](image-url)
shows the temperature dependence of the uncertainty term along with the calculated values from the indirect measurement. The inset a sample. Moreover, the measurement of ΔT_{adb} involves a time span (after application or removal of the magnetic field) where the temperature change remains constant [see Fig. 1(b)] and where there is no significant heat loss. For example, according to Fig. 1(a) there is only a 0.1% temperature difference between the temperature before the start of step 1 and after the end of step 4. The uncertainty involved in this measurement is due to the resolution of the temperature sensor and the small heat dissipation from the sample during the magnetization and measurement process. Considering the heat loss (0.9%/s) from the sample during the time required for the magnetic field (360 ms to achieve 1.8 T with 5-T/s sweep rate) and the time required for the thermocouple response (∼200 ms), an uncertainty of less than 0.5% of the measured value of ΔT_{adb} can be estimated.

IV. DATA ANALYSIS

Fig. 3(a) shows an example of the LCMO compound of the temperature-dependent loop measurement protocol. LCMO exhibits a first-order magnetic transition, i.e., there is a discontinuity in the first derivative of the Gibbs free energy. The first-order nature of the magnetic transition for this compound has been verified using the Arrott plot analysis (the result of the analysis is presented in the Appendix) [26], [27]. During the magnetization process with both positive and negative magnetic fields the temperature of the sample increases, while it decreases during the demagnetization process. In Fig. 3(a), there are temperature shifts between the heating and cooling curves, which is directly related to the temperature hysteresis of the first-order compound. From the temperature-dependent magnetization data (results presented in the Appendix) it is confirmed that there is approximately a 1-K temperature difference in the value of T_C comparing heating and cooling cycles, which causes the temperature hysteresis in the ΔT_{adb} measurement. This temperature hysteresis is an unavoidable consequence of the first-order magnetic transition. Comparing the magnetization and demagnetization curves, there is also a temperature shift, as shown by the orange-colored dotted lines in Fig. 3(a), for an applied magnetic field of ±1 T. This temperature shift occurs due to the fact that the starting temperatures for the magnetization and demagnetization measurements are shifted by ΔT_{adb} (K). For example, as shown in Fig. 3(a), for a magnetic field of 1 T the peak value of ΔT_{adb} = 1.5 K occurs at 250.63 K, and while removing the magnetic field the peak occurs at 252.15 K, which is the sum of the initial temperature and the value of ΔT_{adb} at that temperature. For a first-order magnetic transition there is a shift of T_C toward higher temperature with increasing magnetic field. As a result, the ΔT_{adb} curves shift toward higher temperature with increasing field. Similarly, in the LCMO compound, a temperature shift toward high temperature with the increasing magnetic field has been observed.

The field dependence of the transition temperature T_C has direct consequences on the measurement protocols. In Fig. 3(b), a comparison between the data collected following the continuous cooling and phase reset protocols is presented. Caron et al. [25] have shown that for a first-order compound, the calculated value of the isothermal entropy change following the phase reset protocol is almost half of the value calculated using the continuous cooling protocol. As a reason for this large difference in isothermal entropy change, they pointed out that the basic assumption in Maxwell’s relation

\[
\left(\frac{\partial S}{\partial H}\right)_T = \left(\frac{\partial M}{\partial T}\right)_H
\]

is not valid for first-order magnetic transitions. Fortunately, in the direct measurement, there is no such assumption made and as a result both the continuous cooling and the phase reset protocols indicate the same value of ΔT_{adb}. In addition, there is a small temperature shift between the phase rest and continuous cooling curves, which is also typical for a first-order magnetic phase transition [30].

It is recommended [25] that the phase reset protocol should be adopted for a first-order compound. However, for a real magnetic refrigeration cycle, for example, the active magnetic regenerative (AMR) refrigeration cycle [31], [32], the involved

Fig. 3. (a) Temperature dependent variation of ΔT_{adb} for the LCMO compound for cooling and heating cycles. The different curves correspond to different magnetic fields μ_0H. The positive (negative) values of ΔT_{adb} correspond to magnetization (demagnetization) of the compound. (b) Comparison of ΔT_{adb} measured following continuous cooling and phase reset protocols along with the calculated values from the indirect measurement. The inset shows the temperature dependence of the uncertainty term (T/C)_{unc}.
cooling process is the continuous cooling process. Therefore, while measuring $\Delta T_{\text{ab}}$, data from both protocols should be presented for a basic understanding of the magnetic phase transition as well as for the application of the material in refrigeration cycles.

To compare the direct and indirect measurement data of the MCE, temperature-dependent specific heat measurements at two magnetic fields, 0 and 1 T, have been performed. In the indirect measurement method, the total entropy $[S_H(T)]$ is calculated from the specific heat data by numerical integration of the thermodynamic relation

$$S_H(T) = \int_0^T \frac{C_H(T)}{T} \, dT + S(0)$$

where $S(0)$ is the magnetic field-independent zero-temperature entropy. The $C_H(T)$ data have been collected using the heat capacity option of the quantum design physical property measurement (PPMS) [33]. The calculation is based on the assumption $C_H(0) = 0$, which together with the unknown zero-temperature entropy will introduce a temperature and magnetic field independent constant uncertainty $\delta S_0$. The adiabatic temperature change for the field change $0 \rightarrow H_f$ can then be calculated using the relation [7]

$$\Delta T_{\text{ab}}(T, H_f) = \left[ T(S_H(T) + \delta S_0) - T(S_0(T) + \delta S_0) \right]_S.$$

(7)

Taylor expanding the functions $T(S_H(T) + \delta S_0)$ on the right-hand side of (7), keeping only the first term in the expansion and using the relation $[dT(S)/dT]_H = [T/C(T)]_H$, (7) can be expressed as [7]

$$\Delta T_{\text{ab}}(T, H_f) = \left[ T(S_H(T) - T(S_0)) \right]_S + \delta S_0 \left[ \frac{T}{C_H(T)} - \frac{T}{C_0(T)} \right]_S.$$  

(8)

The second term on the right-hand side of (8) indicates the possible uncertainty involved in the calculation of $\Delta T_{\text{ab}}$. Although the value of $\delta S_0$ is constant, the term $[(T/C_H(T)) - (T/C_0(T))]_S$ is temperature and field dependent; this term will in the following be referred to as $(T/C)_{\text{unc}}$. In Fig. 3(b), the calculated values of $\Delta T_{\text{ab}}$, ignoring the uncertainty contribution from (8), is shown. In the inset of Fig. 3(b), the temperature dependence of $(T/C)_{\text{unc}}$ is shown using $\delta S_0 = 1 \text{ J/kgK}$, clearly showing that the uncertainty involved in (8) will have a positive (negative) contribution on the calculated value of $\Delta T_{\text{ab}}$ below (above) $T_C$. However, Pecharsky and Gschneidner Jr. [7] have pointed out that this uncertainty term can be ignored as the value of $\delta S_0$ is considerably small. Considering a random uncertainty for the indirect measurement yields an uncertainty of about 50% for $\Delta T_{\text{ab}}$ (see the Appendix), while the uncertainty involved in the direct measurement of our system is less than 0.5% as discussed in III. However, the random uncertainty in the indirect measurement can be significantly reduced if the specific heat data are collected with a very small temperature step between measurement points, again at the expense of a considerable measurement time. This reinforces the notion that direct measurements of the MCE are needed for the correct determination of $\Delta T_{\text{ab}}$. Apart from the random uncertainty, there is an experimental limitation with the grease used to mount the sample in specific heat measurements with Quantum Design PPMS. Two greases are typically available for the specific heat measurement, the N-grease that can be used below room temperature and the H-grease that should be used above room temperature. Therefore, materials with a $T_C$ above room temperature require separate measurements with the two greases and each measurement consists of one addenda (background) and one sample measurement. A total of four specific heat measurements is therefore needed to cover the desired temperature range. Noticeably, for the determination of $\Delta T_{\text{ab}}$, the specific heat should be measured for two magnetic fields. This makes a total of eight separate measurements and all measurements should be performed under the same conditions. This makes the indirect measurement challenging for materials with a $T_C$ above room temperature. Similar problems have been encountered for the second-order compound LSMO. The magnetic properties of this compound have been reported elsewhere [23]. Fig. 4(a) shows the temperature dependent variation of $\Delta T_{\text{ab}}$ for this compound measured in the loop protocol. There is no temperature hysteresis (or temperature shift) in the heating and cooling curves, as expected for a

**Fig. 4.** Temperature dependent variation of $\Delta T_{\text{ab}}$ for the second-order materials. (a) LSMO for cooling (open symbols) and heating (filled symbols) cycles and (b) gadolinium (Gd) for cooling, heating, and phase reset cooling cycles. The results for Gd are compared with published data at $\mu_0 H = 1$ T, and 1.5 T, respectively.
second-order material. However, there is a large mismatch between the results from direct and indirect measurements (see the Appendix). This is mostly because of the limitations in the specific heat measurement at higher temperatures that are specific for the, here used, measurement system. Using (6), Pecharsky and Gschneidner Jr. [7] estimated the uncertainty in the entropy calculation and concluded that the uncertainty will be much reduced if the collected specific heat data starts from a very low temperature where the specific heat is negligibly small. Therefore, a material with a high $T_C$ (above room temperature) needs to be measured from a very low temperature, preferably from 2 K or below. In addition, the temperature step used in the specific heat measurement will influence the uncertainty of the calculated entropy. Smaller temperature step yields smaller uncertainty at the expense of a longer measurement time.

However, it should be noted that experimental setups exist that can measure the specific heat accurately [34], [35], as well as specific data processing techniques [36] that yield correct estimates of $\Delta T_{adb}$ from indirect measurements. Still, the requirement of low-temperature data with small temperature steps and background measurements implies long measurement times compared to the direct measurement of $\Delta T_{adb}$.

In order to compare our system with already existing direct measurement systems, a 99.95% pure Gd sample has been measured; the results are shown in Fig. 4(b). Using all the previously described measurement protocols the different results overlap each other, as expected for a second-order material. Our results agree well with the data reported in [37], where a Halbach array magnet and a T-type thermocouple were used as magnetic field source and temperature sensor, respectively. Wang et al. [12] also used a Halbach array magnet, while temperature sensor was an IR sensor. Koskick’ko et al. [38] used a thermocouple as a temperature sensor, while the magnetic field change was realized by moving the sample into the magnetic field. The discrepancy between our results and results reported in [12] and [38] may be related to different purity of the used Gd samples as mentioned by Gimaev et al. [13].

V. CONCLUSION

A simple setup for the measurement of $\Delta T_{adb}$ has been demonstrated here. The $\Delta T_{adb}$ results for the first-order compound LCMO exhibit temperature hysteresis in the heating and cooling measurement process, owing to the temperature-dependent irreversibility of the magnetic phase transition of the compound. The result for $\Delta T_{adb}(H, T)$ from direct measurements has been compared with the indirect result obtained from the specific heat results. Although the results from the two measurements are in good agreement near $T_C$ for the LCMO compound, the large relative uncertainty in the indirect measurement indicates that the direct measurement is superior to the indirect measurement. Apart from the data quality in the indirect measurement, it is also a relatively time-consuming measurement. Moreover, $\Delta T_{adb}$ of a second-order compound (LSMO) has also been presented and the reversibility with respect to temperature of the second-order compound has been verified. We have also studied $\Delta T_{adb}$ for a Gd sample and compared with previously reported results obtained using different types of direct measurement systems.

APPENDIX

The basic magnetic properties of the LCMO compound are presented in Fig. 5. From the temperature-dependent magnetization, a small temperature hysteresis ($\sim 1$ K) is observed. The negative slopes in the Arrott plot analysis confirm the first-order nature of the LCMO compound. The temperature dependence of the total entropy calculated using results from the specific heat measurements are shown in Fig. 5(d) for two different magnetic fields. Within the margin of uncertainty, the change in entropy with the magnetic field is only observed in the vicinity of $T_C$, which explains why both the isothermal entropy change and adiabatic temperature change are zero well below and well above the magnetic phase transition temperature.

In Fig. 6(a), the temperature-dependent values of $\Delta T_{adb}$ for the LCMO compound along with the calculated random uncertainty for the indirect measurement are shown. Following Pecharsky and Gschneidner Jr. [7], the random uncertainty $\sigma \Delta T_{adb}$ can be defined as

$$\sigma \Delta T_{adb}(T, H_f) = \left[ \sigma S_H(T) \frac{T}{C_H(T)} + \sigma S_0(T) \frac{T}{C_0(T)} \right]_S$$

where $\sigma S_0(T)$ and $\sigma S_H(T)$ are the random uncertainties of the total entropy at zero magnetic field and at the magnetic field $H_f$.

Fig. 6(b) shows the measured specific heat using the Quantum Design PPMS system. To calculate $\Delta T_{adb}$ there will be a total of four separate sample measurements and corresponding four separate measurements of the background (addenda). Since the measurement involves the use of two types of grease to hold the sample, it is a challenge to have the same experimental conditions for all eight measurements. As in the case of the LSMO compound, there is a mismatch of specific heat data.
Fig. 6. (a) Temperature-dependent values of $\Delta T_{ab}$ derived from the indirect measurements along with the calculated random uncertainty for the LCMO compound. (b) Specific heat versus temperature for the LSMO compound measured in the PPMS system with N-grease and H-grease at $\mu_0H = 0$ and 1 T. The inset shows $\Delta T_{ab}$ versus temperature obtained from direct and indirect measurements.

between the low-temperature and high-temperature regions. This mismatch creates a large discrepancy [see Fig. 6(b) (inset)] between the direct and indirect measurement results.

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AUTHOR CONTRIBUTIONS

The basic design of the instrument was performed by SG, DH, and PS. DH took a leading role in assembling the hardware components of the instrument. MP has performed the 3-D-drawing of the instrument. Software design, electrical component assembly, calibration of instrument components, sample preparation, data collection, analysis of the results, and writing of the manuscript have been performed by SG. All the authors have read and contributed to the manuscript. The complete work has been performed under the supervision of PS.

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