Evaluation of a long-time temperature drift in a commercial Quantum Design MPMS SQUID magnetometer using Gd$_2$O$_3$ as a standard

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Abstract

The long-time temperature drift in a commercial Quantum Design MPMS SQUID magnetometer was evaluated using time-dependent magnetization measurements of Gd$_2$O$_3$. In contrast to earlier claims, the amplitude of the drift was found not to exceed 1-1.5 K. 30 minutes after system stabilization the temperature deviation did not exceed 0.2 K and the temperature was fully stabilized in less than 3 hours.

Key words: magnetic relaxation, MPMS magnetometer, temperature drift

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1 Introduction

Within last two decades commercial superconducting quantum interference device (SQUID) magnetometers (in a majority of cases Quantum Design MPMS instruments) have truly become the workhorses of many condensed matter physics, chemistry and materials science research and educational laboratories. These instruments are widely used for routine, albeit very accurate, magnetic measurements of wide classes of materials. In the cases of type-II superconductors in the intermediate state or spin-glasses one of the measurements of choice, allowing deeper insight into physical processes, is a time-dependent (relaxation) magnetization measurement [1,2,3,4]. Since a single DC magnetization measurement takes $\sim 30 - 40$ sec, a long total measurement time (several hours) is usually required to have a reliable estimate of the relaxation parameters. In such measurements a knowledge of the long-time temperature drift of the system is important. An estimate of the long-time temperature drift in Quantum Design MPMS-5 instrument was reported by Kopelevich
and Moehlecke [5]. They reported temperature changes as high as \( \sim 4 \text{ K} \) (in 100 - 120 K temperature range) during \( \sim 8 \) hours after the temperature was "stabilized" as indicated by the magnetometer’s own thermometer. Whereas [5] was apparently the first publication to address the temperature drift issue in MPMS instrument, and it sent an important message for the part of the community doing relaxation measurements using this type of magnetometer, the experimental details in [5] were significantly different from the case of routine magnetization measurements, potentially affecting the sample thermalization: the temperature was measured by Pt thermometer mounted in place of a sample. Such a thermometer would, at minimum, require bringing wires to the sample chamber if magnetization probe was utilized or using Quantum Design Manual Insertion Utility Probe (that has a thermal mass significantly higher than the magnetization probe) or its home-made analogue.

In this work we address the issue of long-time temperature drift of a MPMS instrument using time-dependent DC magnetization measurements on \( \text{Gd}_2\text{O}_3 \), a very stable oxide with well-documented [6,7] Curie-Weiss-like temperature-dependent susceptibility that is known to be used for calibration of susceptibility measurements [8]. The use of magnetization measurements for temperature-drift evaluation allows for the experimental protocol to be very similar to routine relaxation measurements and does not alter the thermal mass of the sample assembly.

2 Experimental

\( \text{Gd}_2\text{O}_3 \) powder was obtained from the Materials Preparation Center at Ames Laboratory. The highest magnetic impurities were estimated to be Eu (\( \sim 40 \) ppm) and Fe (< 10 ppm) with total estimated concentration of impurity elements < 120 ppm. To remove possible moisture from the long-time storage, the powder was heated to 400° C for 4 days, cooled down and immediately packed for measurements. The magnetization measurements were made in a Quantum Design MPMS-7 DC magnetometer equipped with a Continuous Low Temperature Control and Enhanced Thermometry (CLTC and ETC) option (M-140) [9] (the scan length was 6 cm, single measurement per point was taken, applied field was \( H = 1 \text{ kOe} \)). 48.3 mg of loose \( \text{Gd}_2\text{O}_3 \) powder (the amount was chosen to ensure reliable signal within the magnetometer’s range at \( 2 \text{ K} \leq T \leq 300 \text{ K} \)) was placed in a gelatine capsule [10]. The remaining capsule volume was filled with \( \sim 50 \) mg of cotton so as to reduce possible motion of the \( \text{Gd}_2\text{O}_3 \) powder. The capsule with the sample was mounted in a transparent drinking straw [11]. Temperature dependent magnetization of the capsule with cotton (background) was measured separately. Over the whole temperature range of interest it was about three orders of magnitude, or more, smaller than total magnetization of the sample and the capsule/cotton together (Fig.
Such accuracy is sufficient for the purpose of this study and in the following, unless stated otherwise, we will refer to the measured magnetization as the magnetization of the sample.

Our temperature-dependent measurements of Gd$_2$O$_3$ show a very clear Curie-Weiss-like behavior (with very small deviations below \( \sim 10 \text{ K} \)) (Fig. 2). These data were taken after stabilizing at 50 K, cooling down to 2 K and taking data on warming, stabilizing temperature every 1 K up to 10 K, every 2.5 K up to 50 K and every 5 K up to 300 K. From the linear fit of the inverse susceptibility, \( \chi^{-1} = H/M \), the Gd$^{3+}$ effective moment was estimated to be \( \mu_{\text{eff}} = 7.86 \pm 0.05 \mu_B \), close to the theoretical value of 7.94 \( \mu_B \) for the Gd$^{3+}$, with the Curie-Weiss temperature of \( \Theta = -16.0 \text{ K} \), similar to the literature data [6,7,8,12].

To analyze the long-time temperature drift of the instrument we will use the Curie-Weiss behavior of the susceptibility of Gd$_2$O$_3$. Since (at least between 10 K and 300 K) the existing data and the current understanding of the magnetic properties of Gd$_2$O$_3$ give no indication of magnetic relaxation processes related to the material, we will assume that any change in the measured magnetization at a nominally fixed temperature is caused by a long-time temperature drift of the sample. To avoid processes related to relaxation of the magnetic field in the superconducting magnet of the instrument, the field was set to 1 kOe in the persistent mode about an hour before the time dependent measurements were started and was not changed during the measurements. We will not use our \( \chi(T) \) data to extract the exact temperature of the sample during such measurements, but rather will look at the evolution of the magnetization and will convert the magnetization drift \( \Delta M \) (taken with respect to the ”stabilized” magnetization after \( \sim 10 \text{ h} \) of measurements) into a temperature drift \( \Delta T \). It should be noted that for convenience in dealing with the experimental data, simple equations below are written for the sample as it is, without a normalization to its mass or molecular weight. The interested reader can use the sample mass and the molar mass to convert these equations into whatever form or units are convenient. We can write the inverse magnetization as

\[
1/M = a + bT
\]  

(1)

The parameters \( a \) and \( b \) are determined from the linear fit of the inverse magnetization. From the Eq. 1 we obtain

\[
\frac{dM}{dT} = -\frac{b}{(a + bT)^2} = -\frac{b}{M^2}
\]  

(2)
Consequently

\[ \Delta T = \Delta M \frac{1}{dM/dT} = -\Delta M \frac{M^2}{b} \]  

(3)

From the \( M(T) \) measurements in \( H = 1 \) kOe the value of \( b \) was determined to be 0.4828 emu\(^{-1}\)K\(^{-1}\).

Two sets of time-dependent magnetization measurements were performed. For each measurement temperature in the first set the sample was heated to 300 K, held there for 10 sec and then cooled to the desired temperature at nominal 10 K/min rate. 10 sec after the temperature was “stabilized” as determined by the magnetometer’s thermometer the time-dependent measurements started. For the second set the sample was first cooled down to 10 K, kept at that temperature for 10 sec and then heated up to the desired temperature at nominal 10 K/min rate. Similarly to the first set, 10 sec waiting time was included before the start of the time-dependent measurements. An example (for \( T_{set} = 50 \) K in the first set of data) of the sequence used for time-dependent magnetization measurements is given below:

Set Temperature 300.000K at 10.000K/min
Wait for Temp: Stable Delay: 10 secs
Set Temperature 50.000K at 10.000K/min
Wait for Temp: Stable Delay: 10 secs
Scan Temp from 50.000K to 50.000K at 3.000K/min in 200 steps (0K/step) Settle
Wait for Temp: Stable Delay: 30 secs
Measure DC: 6.00cm, 48 pts, iscans, AutoRng, Long, Iterative Reg., track: Yes, raw: No, diag: No
End Scan

3 Results

Fig. 3(a) shows the time-dependent temperature drift for \( T_{set} = 100 \) K as measured by the magnetometer’s sensor and as calculated using the Eq. 3 from the changes in measured magnetization of \( \text{Gd}_2\text{O}_3 \) for two sets of measurements - after heating to 300 K (set 1) and after cooling to 10 K (set 2). In both cases the magnetometer’s thermometer does not show any time drift and these data simply indicate the temperature noise tolerated by the magnetometer’s stability algorithm. The temperature drift as inferred from the magnetization drift is \( \sim 0.3 \) K (with temperature increasing with time) in the first set and \( \sim 0.1 \) K (with temperature decreasing with time) in the second set. Different signs of \( \Delta T \) are consistent with the protocol of the measurements in each set. In both sets the temperature is practically stable after \( \sim 3 \) hours and
within 0.1 K (0.1% accuracy) within less than an hour. For lower temperature, $T_{\text{set}} = 50$ K (Fig. 3(b)) the temperature drift is significantly smaller and the temperature is stabilized within $\sim 0.01$ K in less than an hour.

Data for different set temperatures are summarized in Fig. 4. Below 100 K the maximum temperature deviation is less than 0.1 K. At and above 100 K the deviation depends of the cooling/warming history and reaches maximum of $|\delta T| \sim 1$ K at $T_{\text{set}} = 200$ K. These data represent the extreme temperature excursions in our data. If we plot two similar data sets for the $t = 30$ min data (Fig. 4) the deviations from the stabilized temperatures will be $< 0.2$ K. Over the whole temperature range the temperature is fully stabilized after $t \leq 1$ h below 100 K and after $t \leq 3$ h at and above 100 K. Different time-dependent measurements with nominally the same history may have some spread of $\Delta T$ (e.g. 200 K and 300 K for set 2) so the results in Fig. 4 serve only as guidelines.

4 Conclusions

We evaluated the long-time temperature drift in MPMS SQUID magnetometer between 10 K and 300 K using Gd$_2$O$_3$ time-dependent magnetization measurements and well-established Curie-Weiss-like $M(T)$ behavior of this material utilizing the measurement schedule similar to that used for magnetic relaxation measurements in superconductors or magnetic materials. The maximum temperature deviation and its relaxation depend on the measurement’s history and was found to be within 0.1 K below 100 K and higher, up to 1 K above 100 K. Our estimates give lower $|\Delta T|$ values and shortened temperature stabilization times than those reported in Ref. [5]. This may be due to different experimental approaches for such an estimate. Additionally, our measurements were performed on an upgraded version of the MPMS instrument, so if the magnetometer in Ref. [5] did not have the CLTC and ETC option, an improved temperature control in our instrument is probably (at least partially) the reason for its lower maximum temperature deviations. Our results can serve as a guideline for designing measurement’s schedule for magnetic relaxation studies in commercial Quantum Design MPMS magnetometers.

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Fig. 1. Temperature-dependent magnetization of capsule + cotton (background) and the Gd$_2$O$_3$ sample placed in the same capsule and secured by cotton (sample + background) measured in $H = 1$ kOe applied field.
Fig. 2. Temperature-dependent susceptibility, $M/H$, and inverse susceptibility, $H/M$ of Gd$_2$O$_3$ plotted per mole of Gd$^{3+}$. Dashed line - linear fit to the inverse susceptibility. Inset: low temperature part of the inverse susceptibility (symbols) with an extrapolation of the linear fit to the high temperature susceptibility shown as a line.
Fig. 3. Example of time-dependent temperature drift for (a) $T_{set} = 100$ K and (b) $T_{set} = 50$ K as measured by the magnetometer’s sensor and as inferred from the changes in measured magnetization of Gd$_2$O$_3$ for two sets of measurements (see text for details).
Fig. 4. (●, ○) - Difference between initial temperature (10 sec after the temperature was "stabilized" according to the magnetometer’s sensor) and stabilized temperature as inferred from the time-dependent magnetization measurements as a function of set-temperature for two different protocols - set 1 (cooling from 300 K) and set 2 (warming from 10 K); (∗, +) - difference between the temperature after 30 min of measurements and the stabilized temperature.