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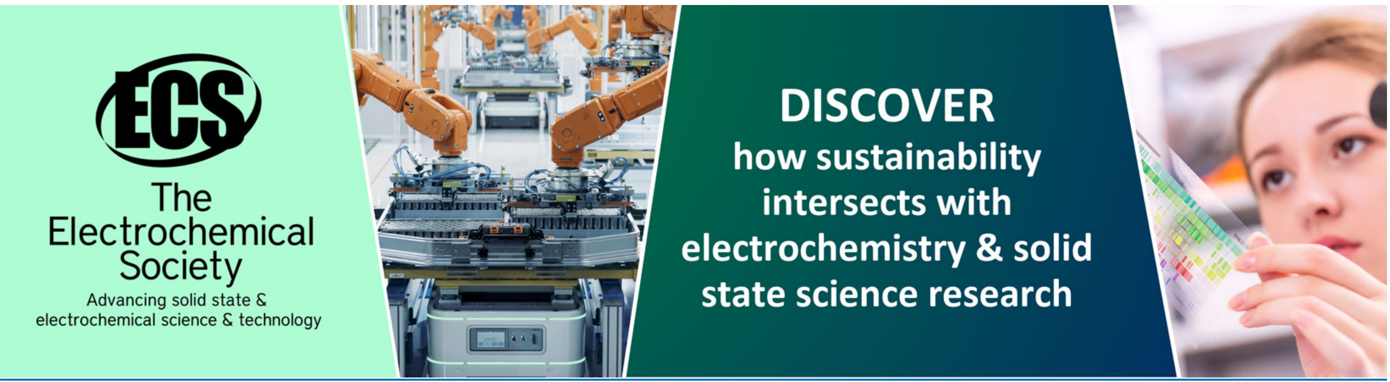

SQUID-based AC magnetometry down to 0.5 K made available on a widely-accessible platform

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Abstract. In order to enable AC magnetometry with the ^{3}He insert that has been designed for Quantum Design's MPMS SQUID magnetometer, we have calibrated the phase shift caused by the insert. Using gadolinium gallium garnet (GGG) as a zero-phase reference, we have succeeded in determining the phase contribution from the ^{3}He insert down to about 0.5 K, below which GGG develops a spin-glass-like transition. This means that AC magnetometry is now feasible down to 0.5 K for frequencies at least up to 100 Hz on the common MPMS platform. At lower frequencies (≤ 1 Hz) virtually no phase shift is added by the ^{3}He insert to the existing shift due to the MPMS sample tube. The sensitivity of the AC magnetization amplitude is estimated to be about 2×10^{-7} emu, although the AC susceptibility of the order of 1×10^{-4} emu shifted by ca. -2×10^{-6} emu when it was measured with the ^{3}He insert, as compared to the normal measurement. This development is expected to facilitate studies of time-dependent magnetic phenomena, such as spin-glass transitions and quantum magnetization tunnelings of single-molecule magnets.

1. Introduction

AC magnetometry is quite useful in investigating time-dependent magnetic phenomena, such as spin glass[1] and magnetic relaxation in superparamagnetic materials, especially in single molecule magnets[2].

Studies of single molecule magnets often require very low temperatures below 2 K[3, 4]. Since the conventional (non-SQUID) AC technique for measuring susceptibility is fairly easy to combine with ^{3}He refrigeration, this kind of apparatuses are not uncommon. SQUID-based AC susceptometer is, however, rather complicated and rarely found, except for Quantum Design's MPMS SQUID magnetometers equipped with the AC susceptibility option. SQUID AC magnetometry has its strength in low frequency measurements thanks to the extremely low-noise nature of SQUID's. But until very recently, the MPMS has had no way of lowering the temperature below about 1.8 K.

One of the authors had successfully developed a ^{3}He refrigeration system for the MPMS[5]. Since then, there has been a question; "Is it possible to measure AC susceptibility down to a ^{3}He temperature region on an MPMS with the AC and ^{3}He options?"

To answer it, we have calibrated the phase shift caused by the ^{3}He insert, using gadolinium gallium garnet (GGG) as a zero-phase reference. We have also checked the frequency dependence of the amplitude.

Up to 200 Hz the phase shift was less than 3.5 ° and the amplitude roll-off due to the attenuation of the AC field was negligible, showing promise for a SQUID AC magnetometry down to 0.5 K. Apart from this, however, the amplitude decreased by about 2×10^{-6} emu even at 1 Hz when the total amplitude was 1.06×10^{-4} emu. The cause of this is still to be worked out.

2. Experimental

The ^{3}He insert for the MPMS has been developed at AIST[5, 6]. ^{3}He gas is liquefied in the double-walled insert made of titanium, situated in the 1.6-K environment the MPMS can provide. After the condensation is complete, the liquid ^{3}He is pumped down to get to low temperature, typically 0.48 K. The sample is immersed in the liquid. DC magnetization measurements are realized by moving the entire insert through the MPMS pickup coil.

The MPMS's AC option enables AC magnetometry by applying a drive field from the modulation coil wound around the SQUID detection coil and picking up the sample's magnetization modulation directly.

Since there are several metal layers between the modulation coil and the sample, the raw measurement has its own phase shift and amplitude dampening. They are measured and corrected for in the MPMS's measurement software. We normally use Dy_2O_3 as a zero-phase-shift reference for this calibration.

In the case of ^{3}He measurements, there are additional titanium tubes inside the modulation coil. They will add some phase shift and will cause some attenuation of both the AC drive field and AC magnetization signal. These systematic errors will have to be measured and corrected for in order to get accurate results.

We have measured the phase shift and the amplitude dampening down to 0.46 K in a frequency range of 0.1–1000 Hz, using gadolinium gallium garnet (GGG) as a reference sample because Dy_2O_3 has some magnetic phase transition around 1 K. GGG also develops some anomaly below about 0.6 K[7], but can cover most of the temperature range relevant to the current study.

3. Results and discussion

When we applied a drive field at 1 kHz around 0.46 K, the base temperature, we found that the maximum AC field we could use without an appreciable temperature rise was 1 Oe. Henceforth we limited ourselves to a 1-Oe modulation at most.

An example AC measurement of the amplitude and phase for one crystal of GGG (the weight unweighed) is shown in Fig. 1. The drive frequency was varied from 1–102 Hz. All the amplitude curves were nearly indistinguishable, ensuring the stability and reproducibility of the AC measurement with the ^{3}He insert. All of them showed a break at around 0.5 K, where the phase at 102 Hz also showed a clear anomaly. This must correspond to the spin-glass transition reported previously[7]. Other than this, the phase at 102 Hz or lower was quite stable and easy to calibrate.

When we raised the frequency to 1 kHz, the phase became around 16 °(Fig. 2). Considering that the MPMS probe itself normally has a phase shift of about 40 ° at 1 kHz, the 16 ° shift may not be surprisingly large. Even so, the phase had a rather large temperature dependence. The plummet at around 0.6 K should be understood as the effect of the spin-glass transition. Apart from that, there was still a significant change of the phase. The phase shift is generally related to some kind of energy dissipation. The insert is made of titanium, whose electrical conductance should be well in residual-resistivity region at these low temperatures and is expected to be fairly constant. It is therefore hard to imagine that the dissipation in the tube walls changed so much. It is very probable that this temperature-dependent phase shift was actually coming from the sample itself. To prove it, we will have to use different paramagnetic materials without a magnetic transition down to very low temperatures.

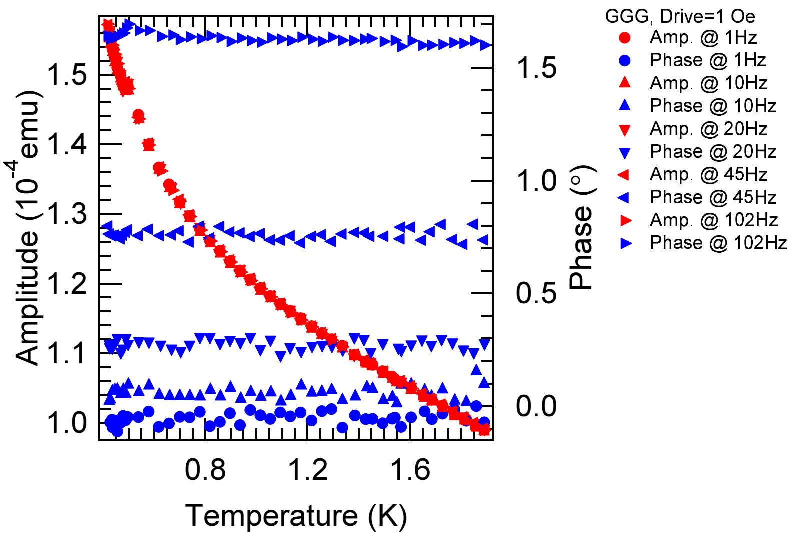

Figure 1. The temperature dependences of the amplitude and phase of a single crystalline GGG at several drive frequencies.

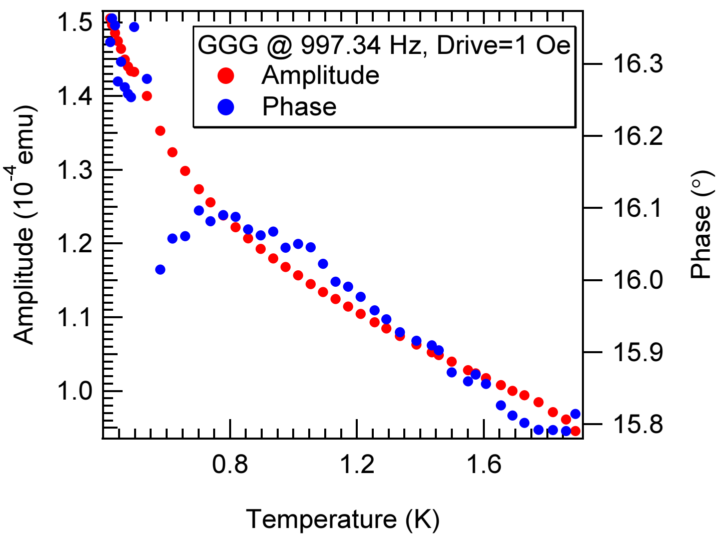

Figure 2. The temperature dependence of the amplitude and phase of a single crystalline GGG at 997-Hz AC field.

Next we evaluate the sensitivity of our AC measurements. As can be seen in Fig. 3, the relative noise increases naturally as the drive field decreases. From this we conclude that amplitudes of $2\text{–}3{\times}10^{-7}$ emu can be resolved.

Finally we make a brief statement about our ongoing study on the accuracy of these AC measurements with the ^{3}He insert. The direct comparison between the data sets with and without the insert can only be made above 1.6 K, to which the MPMS can reach without the help of ^{3}He. The result is shown in Fig. 4. The high-frequency (above 200 Hz) roll-off of the amplitude with the insert is quite reasonable and should be calibratable. Apart from that, however, there is a shift of about -3×10^{-6} emu from the value without the insert, i.e., for the normal measurement. The origin of this shift is still unknown and we are working it out intensively.

In summary we have measured the phase shift and amplitude dampening caused by the ^{3}He insert for the MPMS SQUID magnetometer in AC-susceptibility measurements down to about 0.5 K. Up to 100 Hz the phase shift is less than two degrees and quite stable. Moreover the amplitude roll-off is negligible up to there. At 1 kHz the temperature dependence of the phase shift was rather large, presumably due to the GGG sample we used. These results indicate the

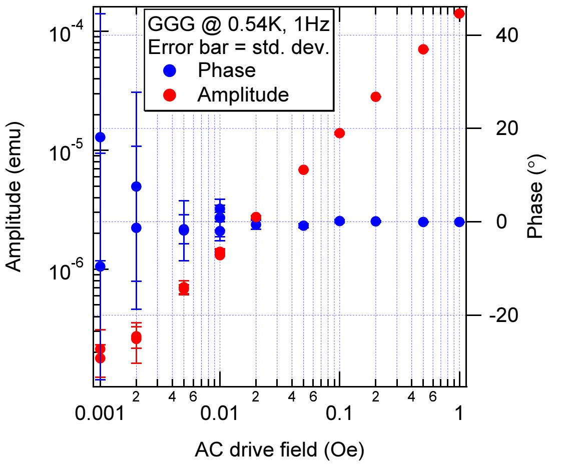

Figure 3. The AC modulation-field dependence of the amplitude and phase of a single crystalline GGG at 0.54 K. The drive frequency was fixed at 1 Hz.

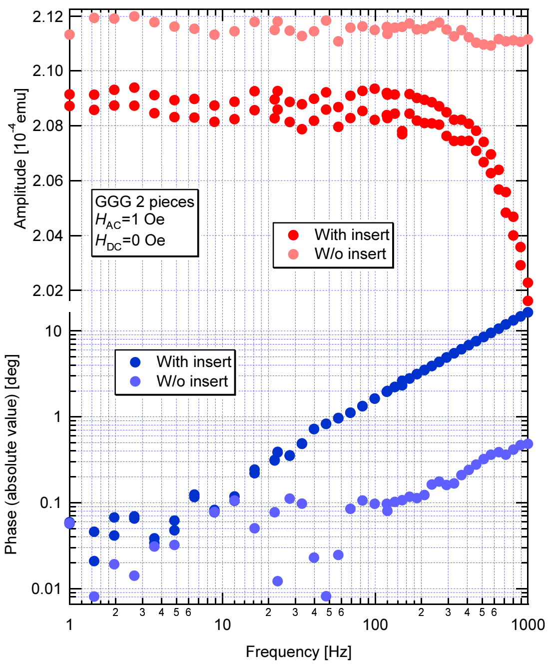

Figure 4. The frequency dependence of the amplitude and phase of a single crystalline GGG at 1.6 K. The red and pink circles denote the data with the insert and without it, respectively. Two curves in red reflects the temperature drift during the two runs. The difference the red and pink cannot be ascribable to the temperature drift.

feasibility of SQUID-based AC magnetometry down to 0.5 K on a popular commercial platform. There is an accuracy problem left to be dealt with.

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