

Assigning an Absolute Scale to Susceptibility Measurements with the ACDR

Background

In a measurement of the dynamic (AC) susceptibility a stationary sample is subjected to a small applied magnetic field which varies (typically sinusoidally) in time. The resulting change in magnetization is probed by measuring the magnitude and phase of the voltage induced in a concentric detection coil. Finally, calibration constants are used to scale this voltage to achieve the reported magnetization in units of moment per field (commonly, emu/Oe). As simply stated, such a measurement can suffer from accuracy problems due to the presence of materials with non-zero AC susceptibilities inside the detection coil, besides the sample, which cannot be removed due to engineering constraints.

A solution which mitigates this issue is to physically translate the sample and perform the measurement at different positions within the excitation/detection coil set. The Quantum Design AC Measurement System (ACMS-II) is able to account for (and automatically remove) background signals and temperature drift-induced errors by leveraging this technique, providing a marked improvement in measurement accuracy. Unfortunately, this multi-point technique is not available when probing AC susceptibility at millikelvin temperatures using the ACDR option; the experiment is limited to the 'single point' configuration, which should be considered only a qualitative measure for the aforementioned reasons. **Figure 1** shows the discrepancy which arises between measuring the same sample with the two different options.

If the user has access to both the ACDR and the ACMS-II options this situation can be remedied. By utilizing the overlap region in temperature between the two options the ACDR data can be corrected, such that it is accurate to within the specified tolerance of the ACMS-II.

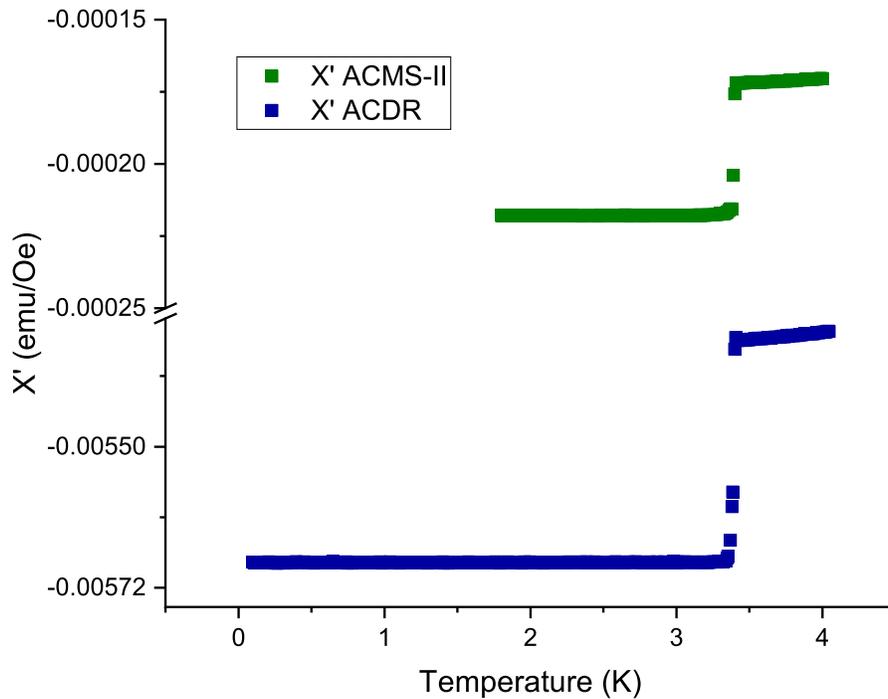


Figure 1: X' values of an indium sample (with geometry matching that of the GGG reference sample included in the ACDR User Kit) measured with both the ACDR and the ACMS-II. Due to the integrated background subtraction, the absolute value of the ACMS-II measurement is considered to be most correct, with the ACDR data exhibiting both a different scale and constant offset.

Comments on Quantities Measured

Consider first a sample's 'true' dynamic (AC) susceptibility which has real (in-phase) and imaginary (out-of-phase) components to its response, indicated by χ' and χ'' respectively (this notation, lacking a subscript, typically implies the volume susceptibility [1]). Note also that the ACMS-II and ACDR options do not report volume susceptibility, but instead the real and imaginary components of the corresponding extrinsic values for a particular sample, given by the Latin characters X' and X'' . For an AC measurement, the volume susceptibility χ and reported quantity X are related by:²

$$\chi' = \frac{1}{V} \cdot X' = \frac{1}{V} \cdot \text{Re} \left[\frac{\partial m}{\partial H} \right] = \text{Re} \left[\frac{\partial M}{\partial H} \right]$$

$$\chi'' = \frac{1}{V} \cdot X'' = \frac{1}{V} \cdot \text{Im} \left[\frac{\partial m}{\partial H} \right] = \text{Im} \left[\frac{\partial M}{\partial H} \right]$$

...where the volume magnetization is indicated by the typical symbol M and is related to the net magnetic moment of the sample m by

² The stated equivalence to the differential expressions is generally valid only in the limit of low frequencies for non-hysteretic materials where $\chi'' \rightarrow 0$.

$$M = \frac{m}{V}$$

Since a precise measure of the volume is often impractical for samples of the size usually employed, often a more convenient intrinsic quantity used is the mass susceptibility χ'_ρ :

$$\chi'_\rho = \frac{1}{\rho} \cdot \chi' = \frac{1}{b} \cdot X'$$

...where the sample density is ρ and its mass is b , and the analogous form is implied for χ''_ρ .

This is mentioned here to stress the relation between the extrinsic quantity reported by the QD measurement options (X'), and the intrinsic quantities most often of interest for purposes of comparison or use in further calculations (χ', χ'_ρ). The remainder of the treatment here deals exclusively in terms of X' and X'' with the understanding that a simple multiplicative scaling by volume, mass, or number of moles will still generate the corresponding intrinsic values.

Relation Between ACMS-II and ACDR Data

When measuring AC susceptibility in a gradiometer-based system there are generally two main corrections to the reported value which need to be considered: a constant offset term (D) arising from any background signal present, and a multiplicative scale factor (C) which accounts for the particular spatial distribution of a real sample. These correction terms relate the raw measured quantity ($X'_{\text{meas.}}$) to the 'true' value (X') as follows:

$$X' = C \cdot X'_{\text{meas.}} + D$$

With the ACMS-II the sample can be moved to various points within the gradiometer, allowing for automated subtraction of the constant background term's contribution [2]; the scale factor C applied by the ACMS-II option software is calibrated at the factory using a cylinder of GGG included in the User Kit.⁴ If the sample in question has geometry similar to that of the GGG cylinder, it is a reasonable assumption that the reported value $X'_{\text{meas.}}$ is equivalent to the real property X' to within the specifications of the option.

When the sample form-factor differs from the GGG cylinder significantly, it may be necessary to account for this deviation [3], [4]. It is left to the user to determine if any potential correction to the factory-set scale factor is significant or not [5], [6]; any accounting for this must be done outside of MultiVu after the initial measurement.

Contrast this with the ACDR where instead the *ad hoc* nulling procedure is designed to remove the bulk of the contribution of the D term from the signal. This technique is still less-than-ideal though, compared with the multi-point ACMS-II measurement, and for individual samples with small dynamic susceptibility signals the remaining background component of the total signal may still be significant.

⁴ C is chosen such that the resultant real component X' matches the slope of the DC moment m vs. DC applied field H for the GGG cylinder; the DC moment is itself calibrated earlier to a known Pd reference.

Since the ACDR and ACMS-II coil sets differ significantly in their construction their respective scale factors are not be the same. This is due to the scale factor's dependency on the details of gradiometer (mainly: coil diameter, spacing between the sets of turns of wire, and number of turns in each set). Consider then the general case of the same sample (of arbitrary shape) measured in both the ACDR and the other in the ACMS-II:

$$X' = C_{ACDR} \cdot X'_{ACDR} + D$$

$$X' = C_{ACMS} \cdot X'_{ACMS}$$

Where $X'_{[OPTION]}$ indicates the value reported by MultiVu for the indicated measurement option (implicitly understood to be a function of temperature, field, and frequency), and $C_{[OPTION]}$, D , and X' are all unknown. By measuring identically the same sample⁹, the right-hand-sides of both expressions can be equated such that:

$$C_{ACMS} \cdot X'_{ACMS} = C_{ACDR} \cdot X'_{ACDR} + D$$

...simplifying this:

$$X'_{ACMS} = C_T \cdot X'_{ACDR} + D_T$$

$$\dots \text{ where: } C_T = \frac{C_{ACDR}}{C_{ACMS}}, D_T = \frac{D}{C_{ACMS}}$$

This equality holds for an individual ordered pair (X'_{ACDR} , X'_{ACMS}) collected at the same temperature, applied DC field, AC excitation field amplitude, and excitation frequency. Plotting the two reported X' values against each other across the region of temperature overlap should reveal a linear relationship between the two; a fit of this line will provide values for C_T (the slope) and D_T (the y-intercept), as shown with the example data in [Figure 2](#). With these two quantities the ACDR dataset can be properly mapped to the ACMS-II set, as shown in [Figure 3](#), extending the absolute accuracy of the former down to the DR base temperature of 50 mK.

If the details of the sample geometry and gradiometer coil configuration are treated theoretically and a value for either C_{ACMS} or C_{ACDR} is derived, then additionally the other $C_{[OPTION]}$ quantity can be determined alongside D , as well as the 'true' X' . The process must be repeated for the X'' quantities as the scale and offset values are not identical.

⁹ If it is impractical to measure the same sample with each technique, samples of nominally identical material (which would have the same χ'_p) can be used. If the mass b of each is known, analysis similar to what follows may still be completed upon substituting $b_{[OPTION]} \cdot \chi'_p = X'$, where now the intrinsic mass susceptibilities can be directly equated between the two expressions.

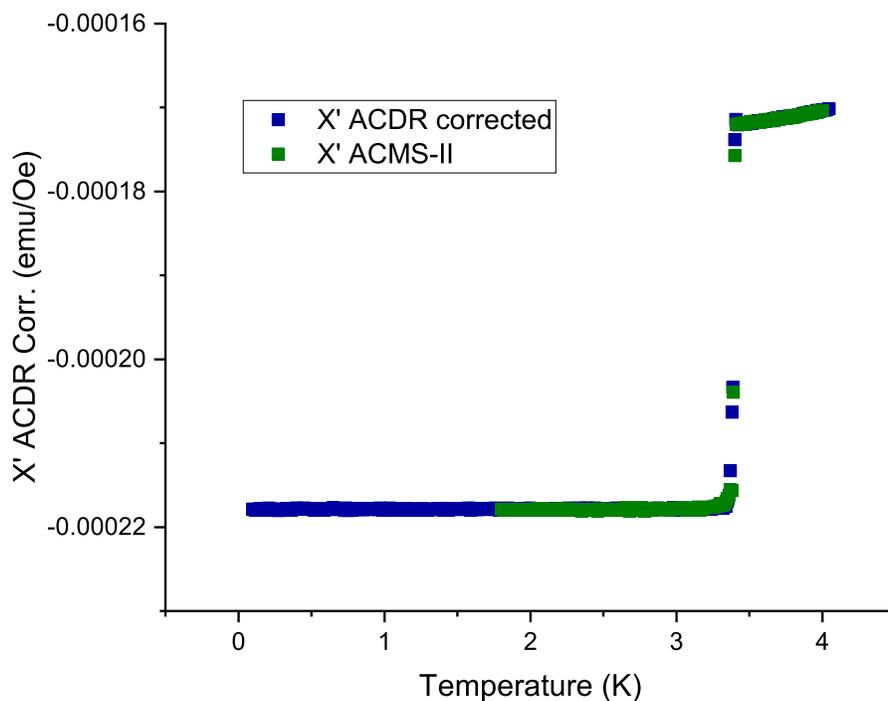


Figure 3: X'_{ACDR} values, corrected using the extracted fit parameters, plotted alongside the initially reported values of X'_{ACMS} , first shown in [Figure 1](#). The ACDR values, extending down to 50 mK, are now accurate to within the stated specifications of the ACMS-II option.

Experimental Considerations

Measuring the Sample Using the ACDR

Take care when measuring with the ACDR to ensure a proper, successful nulling is collected before installing the sample (this will minimize the remaining offset component D of the reported signal). Confirm that the measurement range covers at least the full range of overlap between the two options (1.9 K – 4.0 K). Also take note of the excitation amplitude and frequency- it is best if these are as closely matched between the two options/measurements as possible since generally the dynamic susceptibility is a function of these parameters.

Recall as well that the ACDR affords much smaller AC field amplitudes (4 Oe maximum) than the ACMS-II (15 Oe maximum). The two options allow identical ranges of excitation frequencies, but measurement sensitivity experiences a roll-off, decreasing an order of magnitude with every decade's decrease in drive frequency. It is also worth restating that for both options, when accuracy/precision is of particular importance the recommended mode of measurement is to only collect data when the temperature/field is stable (as opposed to scanning/sweeping).

Measuring the Sample Using the ACMS-II

As before, attempt to match the experimental conditions as closely as possible (excitation amplitude and frequency, stabilization conditions at each temperature). A sufficiently high point density in the overlap region is also crucial, particularly if the sample response varies strongly with temperature (ie: phase transitions), since only the points in the overlap will be useful in the linear fit to determine the scaling and offset values.

Since the two options use incompatible sample holders, take care when re-mounting from one to the other to preserve the sample's orientation to the extent possible. Regardless of if the same sample or a different one (of the same material) is used, a careful accounting of the actual sample mass present in each measurement is important to the final accuracy of the combined dataset if the scale factor is to be independently calculated.

Note on Temperature Accuracy

The specified accuracy of the DR thermometry depends on the temperature ($\pm 1\%$ at 4 K; $\pm 2\%$ at 300 mK; $\pm 10\%$ at 50 mK), whereas the ACMS-II temperature sensor is calibrated using the PPMS sample chamber thermometer, which itself has a specified accuracy of $\pm 1\%$. It is conceivable, and in fact probable, that a small offset in the temperature-dimension may exist between data collected using the ACDR and the ACMS-II, even after properly scaling the ACDR values using the above technique, despite both options meeting their stated specifications.

Typically this is on the order of 10's of mK, and for 'complete overlap' one set may need to be shifted by this temperature offset relative to the other. The choice of which to shift is arbitrary, as it corresponds to the uncertainty in the temperature scale intrinsic to both measurements (absent some well-characterized external reference).

For questions or further assistance with this analysis, contact apps@qdusa.com.

References

- [1]** R. B. Goldfarb and F. R. Fickett, "Units for Magnetic Properties - NBS Special Publication 696," U.S. Department of Commerce, National Bureau of Standards, Boulder, CO, 1985.
- [2]** Quantum Design, Inc., "AC Measurement System (ACMS-II) Option User's Manual," 2017.
- [3]** Quantum Design, Inc., "Application Note 1500-015: Accuracy of the Reported Moment- Sample Shape Effects," 2010.
- [4]** Quantum Design, Inc., "Application Note 1500-015: Accuracy of the Reported Moment- Sample Shape Effects," 2010.
- [5]** P. Stamenov and J. M. D. Coey, "Sample size, position, and structure effects on magnetization measurements using second-order gradiometer pickup coils," *Review of Scientific Instruments*, vol. 77, no. 1, p. 015106, 2006.
- [6]** M. E. Hayden, V. Lambinet, S. Gomis and G. Gries, "Note: Evaluation of magnetometry data acquired from elongated samples," *Review of Scientific Instruments*, vol. 88, no. 5, p. 056106, 2017.