AC Susceptibility Measurement in High Frequency Region up to 10 kHz using a SQUID Magnetometer MPMS

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We performed AC magnetic susceptibility measurement for high frequency (f) range, based on a superconducting quantum interference device (SQUID) magnetometer, MPMS (Quantum Design Inc.). Original system of MPMS uses an A/D converter with 12 bit or 16 bit, and there the frequency of AC field is limited to less than 1 kHz by a software. In the present system, we transformed an analog signal of SQUID output to a digital one via an A/D converter with 24 bit, and generated AC field by a signal generator outside MPMS. The maximum f was enlarged from 1 kHz to 10 kHz. However, it has been confirmed that amplitude of AC field (HAC), at which the rf-SQUID works as a current/voltage transformer, decreases with increasing f. The value of HAC was restricted below 0.02 Oe at f = 10 kHz. The performance of the present system has been confirmed via the measurements of a paramagnetic material Dy2O3 and a rare-earth ferromagnet Ho. The normalization constant for the magnitude of the SQUID output, to calibrate the effect of eddy current, is presented as a function of frequency.

1. Introduction

Nowadays the magnetic property measurement system, MPMS (Quantum Design Inc.), is a popular magnetometer-susceptometer, and the transformation of magnetic flux into voltage is performed by rf type of a superconducting quantum interference device (SQUID) [1]. SQUID is generally a current-voltage transformer, which works without AC magnetic field. In MPMS, we can measure a magnetization at high DC magnetic fields as well as an AC magnetic susceptibility. Its performance for the AC susceptometer is guaranteed for frequencies of \( f \leq 1 \) kHz and at low amplitude of AC field (\( H_{AC} \)) (usually 4.0 Oe at maximum). On the other hand, an electro-magnetic induction method detects induced voltage by vibrating a detection coil or measurement sample, or applying AC field. In the AC measurement, its sensitivity increases with increasing \( f \) and \( H_{AC} \). As a commercial system using the above method, the physical property measurement system, PPMS (Quantum Design Inc.) is well known, and it covers the frequency region of \( f = 10 \) Hz ~ 10 kHz.

Recently, nano-size magnets such as a magnetic nano-particle and a single-molecule magnet have attracted much attention of material scientists. In order to understand these magnetic properties in detail, we need the AC magnetic measurement over the wide range of frequency as well as the DC magnetic measurement at low temperature and high DC field. However, it is difficult to perform both magnetic measurements in one apparatus. In principle, MPMS utilizes the rf-SQUID, which works for
We modified some factors in the AC magnetic measurement system of MPMS, so that we enlarged the frequency range of AC measurement up to 10 kHz.

2. Experimental

Prior to describing the present system, several factors in the conventional AC measurement using MPMS are mentioned. The A/D converter for the SQUID output ($V_S$) has 12 bit resolution. The frequency of AC field is restricted below 1 kHz by a software, MultiVu. Between the sample space and NbTi detection coil, three layers of quantalloy are placed. Therefore the effect of eddy current cannot be ignored, especially at high $f$.

In the modified system, we use the MultiVu for the operations except for applying AC field and detecting $V_S$ from the rf-SQUID system. The overview of the present system is shown in Figure 1. When high $f$ of above 1 kHz was applied, we must use a signal generator outside MPMS, ex. FG120 (Yokogawa Electric Corporation). The analog signal of $V_S$ passed through a filter in MPMS was digitized by a 24 bit A/D converter with sampling rate $f_s$ of 50 kHz at maximum (NI9233, National Instrument Inc.), which functions effectively in acoustic frequency region of from 2 Hz to 20 kHz. NI9233 was connected via USB2.0 (universal serial bus) to a computer, in which a LabVIEW program (National Instrument Inc.) was operated. The digital signal of $V_S$ was analyzed by the Fourier transformation included in the LabVIEW program. The information of temperature ($T$) obtained by MultiVu was sent to the LabVIEW program through TCP/IP. The signal generator was connected with the computer via GPIB (general-purpose interface bus), and could be controlled by the LabVIEW program.

A series of procedure in the actual measurement is as follows: $T$ is varied according to a sequence program of MultiVu, which is prepared prior to starting the measurement. We obtain the information of $T$, every two seconds, via TCP/IP. At that time, the sample is located in the center of the detection coil system. When $T$ reaches a certain target temperature, AC field is applied. And, after ten seconds, the reference signal ($V_r$) and $V_S$ digitized by NI9233 are taken in the computer. The number of sampling is usually five thousand in the condition of $f_s = 50$ kHz. At the present condition, the value of $H_{AC}$ is estimated according to the following relation, $H_{AC} = A \times V_r / (R^2 + (L \times 2\pi f)^2)$, where $R$ and $L$ are the resistance and inductance in the coil line for applying AC field, respectively. Here the coefficient $A$ is estimated by the calibration experiment using a Hall sensor. In the case of complex Fourier transformation, the determination of the orthogonal coordination system prior to the action is important. In the present system, the determination has not been trustworthy yet because of insufficiency in the quantity of experimental data. Herein we mainly focus our attention on the variation in the amplitude of $V_S$.

![Figure 1](image.png)

**Figure 1.** Overview of our AC measurement system, based on MPMS. $V_r$ is the voltage of the reference signal, $V_S$ is the voltage of the SQUID output and $T$ is the temperature. The refrigerator of MPMS includes a series of rf-SQUID system (SQUID device, the controller, and detection coils), AC field coil, thermometers, heaters, and so on.
3. Experimental results

Figure 2 shows frequency dependence of $V_S$, amplified by an amplifier in the rf-SQUID system at $T = 20$ K in the measurement of a molecule-based ferrimagnet with a transition temperature of 37 K [2]. Indeed, $V_S$ includes the background contribution as well as the sample contribution, and the frequency dependence reflects the eddy current effect on each contribution in $V_S$. In the frequency region below 1 kHz, we could apply the AC field of 4.0 Oe. For $f > 1$ kHz, however, the permissible limit of $H_{AC}$ decreases with increasing $f$. For instance, the maximum value is 0.02 Oe at $f = 10$ kHz. The horizontal axis of Fig.2 represents $V_S$ normalized with $H_{AC}$. The flat frequency dependence was confirmed for $f \leq 5$ kHz, and the contribution of the background signal in $V_S$ increases with increasing $f$, especially for $f > 5$ kHz. Namely, even below 5 kHz, the signal of a target material must decrease with increasing $f$. In MPMS, three layers of quantalloy are inserted inside both the AC field coil and the detection coil, and the eddy current effect is inevitable. In addition, a noise peculiar to the rf-SQUID exists at around $f = 20$ kHz. $f_s$ of NI9233 is 50 kHz, and for $f > 10$ kHz, the separation of $V_S$ form the huge noise of 20 kHz was difficult. If an A/D converter with higher $f_s$ as well as high bit (ideally 24 bit) could be utilized, the frequency range of the AC magnetic measurement would be further enlarged, causing the upper limit of frequency range to approach 20 kHz.

![Figure 2](image-url)

**Figure 2.** Frequency dependencies of $V_S/H_{AC}$ amplified by the amplifier in the rf-SQUID system, at $T = 20$ K in the measurement of a molecule-based magnet [2]. The range and gain in the amplifier were $\times 100$ and $\times 2$, respectively.

![Figure 3](image-url)

**Figure 3.** Temperature dependences of AC susceptibility for Dy$_2$O$_3$ at $f \leq 10$ kHz. The range and gain in the amplifier were $\times 1$ and $\times 2$, respectively. Then, 1.0 V/Oe in $V_S/H_{AC}$ corresponded to $7.0 \times 10^{-8}$ emu. The inset shows the frequency dependence of normalization factor $N$.

Figure 3 shows the AC magnetic susceptibility of Dy$_2$O$_3$ as a function of $T$ at several frequencies. The applied $H_{AC}$ varied as follows: $H_{AC} = 0.07$ Oe for $f = 40$ Hz and $300$ Hz, $H_{AC} = 0.066$ Oe for $f = 1$ kHz, $H_{AC} = 0.035$ Oe for $f = 5$ kHz and $H_{AC} = 0.019$ Oe for $f = 10$ kHz. There $V_S$ is normalized with $H_{AC}$, and afterward the unit of AC susceptibility was converted from V/Oe to emu, based on the same measurement at $f = 40$ Hz and 1 kHz by the conventional way. The mass of the sample was 144 mg. According to comparison with the data by conventional way, the contribution of the subtracted background signal against the $V_S$ was estimated to be 69% at $f = 40$ Hz, 70% at $f = 300$ Hz, 70% at $f = 1$ kHz, 85% at $f = 5$ kHz, and 94% at $f = 10$ kHz. Dy$_2$O$_3$ is a prototype of paramagnetic material. It has been known that the out-of-phase does not appear, and the amplitude of the in-phase does not vary against the change of $f$ [3]. The normalization constant ($N$) was estimated so that the amplitude of AC susceptibility at each $f$ becomes equal each other, and the frequency dependence of $N$ is shown in the inset of Fig. 3. $N$ exhibits almost linear frequency dependence.
We measured the AC susceptibility for Ho, which has the ferromagnetic order at around 16 K. The mass of the sample was 250 mg. Figure 4 shows the AC susceptibility for Ho as a function of $T$ at $f = 40$ Hz, 1 kHz, 5 kHz and 10 kHz. The applied $H_{AC}$ varied as follows: $H_{AC} = 0.07$ Oe for $f = 40$ Hz, $H_{AC} = 0.03$ Oe for $f = 1$ kHz, $H_{AC} = 0.03$ Oe for $f = 5$ kHz and $H_{AC} = 0.007$ Oe for $f = 10$ kHz. The AC susceptibility was multiplied with the normalization factor shown in the inset of Fig. 3. At all frequencies, the distinct magnetic anomaly due to the ferromagnetic order was observed, and the magnitude hardly changed. Herein, each value at $T = 60$ K was shifted every $1 \times 10^{-3}$ emu. We have confirmed that these results at $f = 40$ Hz and 1 kHz, in which the conversion of the unit form V/Oe to emu has been performed, were consistent with those by the conventional way.

We have succeeded in detecting the AC magnetic susceptibility in the frequency region up to 10 kHz, by keeping $H_{AC}$ in low level and multiplying appropriate $N$. However, there remain some technical items to be solved at present: At first, when the sample is measured in the detection coil system and the subtraction between the signals at two positions is performed, the effective subtraction of background signal for the total $V_S$ is successful. Next, the time delay of $V_S$ relative to $V_r$ will be determined precisely, the complex Fourier transformation could be performed. Finally, if $f_s$ will increase, the experimental accuracy on signal analysis for $f > 5$ kHz would be enhanced.

![Figure 4. Temperature dependencies of $V_S/H_{AC}$ of Ho at $f = 40$ Hz, 1 kHz, 5 kHz and 10 kHz. The range and gain in the amplifier were $\times 10$ and $\times 5$, respectively. Then, 1.0 V/Oe of $V_S/H_{AC}$ corresponded to $3.0 \times 10^{-7}$ emu.](image)

4. Conclusion

We performed AC magnetic susceptibility measurement for frequency range up to 10 kHz, based on the SQUID magnetometer MPMS. We transformed an analog signal of SQUID output to a digital one via an A/D converter with 24 bit, and AC field was generated by a signal generator outside MPMS. The AC susceptibility for $f \geq 5$ kHz could be observed by keeping the amplitude of AC field ($H_{AC}$) in low level. $H_{AC}$ had to be decreased with increasing $f$, and the value was restricted below 0.02 Oe at $f = 10$ kHz. In order to evaluate the absolute value of the AC susceptibility, the normalization constant for calibration of the eddy current was estimated via the measurement of the paramagnetic material Dy$_2$O$_3$, and the validity of the system was confirmed through the measurement of rare-earth ferromagnet Ho.

References

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