Development of integrated AC-DC magnetometer using high-$T_c$ SQUID for magnetic properties evaluation of magnetic nanoparticles in solution

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Abstract. We developed an integrated AC-DC magnetometer using a high critical temperature superconducting quantum interference device (high-$T_c$ SQUID) to evaluate the static and dynamic magnetic properties of magnetic nanoparticles (MNPs) in solution. The flux-transformer method consisted of first-order planar and axial differential coils that were constructed for static and dynamic magnetization measurements, respectively. Vibrating-sample and harmonic detection techniques were used to reduce interference from excitation magnetic fields in the static and dynamic magnetization measurements, respectively. Static and dynamic magnetization measurements were performed on commercially available iron oxide nanoparticles in diluted solutions. The magnetic responses increased with the increase in concentration of the solutions in both measurement results. The magnetization curves showed that the diamagnetic signal due to the carrier liquid of the iron oxide nanoparticles existed in a dilute solution. Biasing with a proper DC magnetic field in the dynamic magnetization measurement resulted in improved signals of the second and third harmonics. Therefore, highly sensitive magnetic characterizations of MNPs utilizing the static and dynamic magnetization measurement are possible via the developed system.

1. Introduction
Magnetic nanoparticles (MNPs) are attracting interest in fields such as bio-immunoassay, magnetic nanoparticle imaging, and magnetic drug targeting due to their promising results on a nanometer scale. In order to detect MNPs, their inherent magnetic properties are measured by magnetic remanence [1], relaxation [2] and susceptibility methods [3]. Measurement systems that utilize the former two methods can achieve high sensitivity due to less/no interference from excitation magnetic fields. However, magnetic susceptibility requires measurement in the presence of excitation magnetic fields, thus, the interference from these fields will limit the sensitivity, particularly in AC systems. Harmonics generated from the nonlinear magnetization characteristics of MNPs have been utilized to reduce the interference of excitation magnetic fields in the dynamic magnetization measurement [4]. These harmonics are used to quantify MNPs with fast measurements and improve sensitivity by isolating the frequency component of excitation magnetic fields. Large amplitudes of excitation magnetic fields, which can cover wide regions of the nonlinear characteristics, are necessary to

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increase the generation of harmonics. In the static magnetization measurement, high sensitivity is achieved using the vibrating-sample technique and a superconducting quantum interference device (SQUID) with a flux-transformer [5].

The combination of the static and dynamic magnetization measurement techniques in a magnetometer system can reveal further information for analysis of MNPs in solutions. With the aim of achieving a compact, low-running cost and highly sensitive magnetometer to evaluate the magnetic properties of magnetic nanoparticles in solutions, we developed an integrated AC-DC magnetometer using a high critical temperature superconductor (high-$T_c$) SQUID with a flux transformer method based on our previously developed system [6]. For measurements of static and dynamic magnetizations, different coil arrangements were adopted to increase the sensitivity. To demonstrate the sensitivity of the system, we measured the magnetic properties of iron oxide nanoparticles in solutions with respect to different concentrations.

2. Experimental

2.1. Measurement system configuration

An overview of the developed system and coil arrangements for static and dynamic magnetization measurements is shown in figure 1. We employed first-order planar and axial differential coils for static and dynamic magnetization measurements, respectively, as the pickup coils in order to reduce the environmental noise being transferred to the SQUID. Both types are constructed from identical elliptical coils with two coils connected in a series-opposing configuration. The external major and minor axes, internal major and minor axes, length, resistance, inductance and Cu wire diameter of one elliptical coil were 26 mm, 13.5 mm, 15 mm, 1.5 mm, 3.9 mm, 2.99 $\Omega$, 451.6 $\mu$H, and 0.32 mm, respectively. The baselines of the planar and axial differential coils were 13.5 mm and 18 mm, respectively.

In the static magnetization measurement, a sample was exposed in a DC magnetic field generated by an electromagnet and vibrated perpendicularly to the axis of the DC magnetic field using an actuator. The sensitive axis of the planar differential coil was parallel to the axis of the DC magnetic field as shown in Fig. 1 (a). The induced signals were transferred to the inductively coupled SQUID and the SQUID output was lock-in detected. The motion of the actuator detected by a laser positioning sensor was used as a reference signal at a lock-in amplifier.

![Figure 1](image_url)

**Figure 1.** Schematic diagram of the developed system with coil arrangements of (a) first-order planar differential coil and (b) first-order axial differential coil for static and dynamic magnetization measurements, respectively.
In the dynamic magnetization measurement, the sensitive axis of the axial differential coil was placed perpendicular to the excitation magnetic field so that interference of the excitation magnetic field could be reduced. The sample was placed at the center of the axial differential coil as shown in figure 1 (b). An AC magnetic field superimposed with a DC magnetic field was applied to the sample and the induced signal, which was amplified at the SQUID, was lock-in detected using a reference signal at a function generator. The excitation magnetic fields were measured using a Hall sensor.

In this study, we used a ramp-edge type SQUID. The SQUID was fabricated on an MgO substrate using a multilayer fabrication technique. SmBa$_2$Cu$_3$O$_y$ (SmBCO) and Er$_{0.95}$La$_{0.1}$Ba$_{1.95}$Cu$_3$O$_y$ (L1ErBCO) were used as the base and counter electrodes, respectively. The input coil substrate was placed over the high-$T_c$ SQUID gradiometer substrate with an overall size of $7.5 \times 15$ mm$^2$, and their mutual inductance was calculated as being 1.88 nH. Details of the SQUID can be found in [6].

2.2. Solutions of iron oxide nanoparticles

The dilute solutions of iron oxide nanoparticles (nanomag®-D-spio) were prepared by suspending iron oxide nanoparticles in purified water. The diameter of iron oxide nanoparticles amounts to 100 nm and consists of dextran iron oxide composites. Iron oxide concentrations of the prepared solutions were 24 µg/ml, 48 µg/ml, 72 µg/ml and 96 µg/ml, and the solutions were encased in $10 \times 30 \times 10$ mm$^3$ of acrylic cases, respectively.

3. Results and discussion

3.1. Static magnetization measurement

The measurement results of the static magnetization of iron oxide nanoparticle solutions in the range of $-260$ to 260 mT with 20-mT intervals are shown in figure 2. The magnetization curves showed the superparamagnetic characteristic with the saturation magnetic field $H_s$ of approximately $\mu_0H_s = 40$ mT, where $\mu_0$ is the vacuum permeability and $H$ is the magnetic field. The curves responded almost linearly with the change of concentrations. The diamagnetic susceptibility was observed in the magnetic field region higher than 40 mT, indicating the diamagnetism of water as the carrier liquid. This result suggests that the diamagnetic signal from the carrier liquid should be taken into account when measuring dilute solutions of MNPs. The high sensitivity of the developed system showed that it was able to measure the iron oxide nanoparticles in low-concentrated solutions.

![Figure 2. Magnetization curves of iron oxide nanoparticles solutions in different concentrations.](image)

3.2. Dynamic magnetization measurement

To cancel the circumference noise, subtractions between lock-in detected signals of with-sample and without-sample measurements were performed in the dynamic measurement technique. Figure 3 shows the characteristics of second and third harmonics of the 24-µg/ml iron oxide nanoparticles solution with respect to the different AC magnetic field amplitudes biased with the DC magnetic fields.
from 0 to 100 mT. The frequency of the AC magnetic field was 5 Hz. The signals of second and third harmonics increased with larger amplitudes and a proper bias of the DC magnetic field. Figure 4 shows the second and third harmonics at 20 and 0 mT of the DC magnetic field, respectively, during the excitation of 30-mT amplitude of the AC magnetic field. The second and third harmonics responded linearly with the increasing concentrations. The results showed that highly sensitive dynamic magnetization measurement was possible using the developed system.

4. Conclusion
We developed an integrated AC-DC magnetometer using a high critical temperature superconductor (high-$T_c$) SQUID with a flux transformer method for the magnetic properties evaluation of MNPs in solutions. The coil arrangements of the planar and axial differential coils were used for the static and dynamic magnetization measurement techniques, respectively. Using commercially available iron oxide nanoparticles to demonstrate the developed system, high sensitivity was shown in both techniques. Highly sensitive magnetic characterizations of MNPs utilizing the static and dynamic magnetization measurement were possible via the developed system.

**Figure 3.** Second and third harmonic signals of the 24-µg/ml iron oxide nanoparticles solution with respect to the different DC and AC magnetic fields.

**Figure 4.** Second and third harmonic signals upon excitation of 30-mT amplitude of the AC magnetic field with the DC biases of 20 and 0 mT, respectively.

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