Optimization of Detection Unit of AC/DC High-T_c SQUID Magnetometer for Evaluation of Magnetic Nanoparticles in Solution

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***Abstract*— We have optimized the detection unit of the previously developed AC/DC high-Tc SQUID magnetometer and evaluated the magnetic properties of magnetic nanoparticles in solution. A compensation coil technique was used in the fabrication of the detection coil to reduce the interference of the excitation magnetic field. This technique had resulted in a single detection coil using one SQUID for AC and DC magnetization measurement functions and reduced the spatial limitation of the sample shape. The small compensation coil was manually tuned to reduce the interference. Improved sensitivity for evaluation of magnetic nanoparticles can be expected using this technique.**

I. Introduction

MAGNETIC nanoparticles have been studied in various applications, e.g., bio-immunassay [1], [2], magnetic nanoparticle imaging [3] etc, due to their promising characteristics. In applications using magnetic nanoparticles as sensing targets, the measurement performance is mostly influenced by the magnetic characteristics of the magnetic nanoparticles [4]–[6]. Their magnetic characteristics were utilized in applications using magnetic susceptibility [7]–[9], relaxation [6], [10], [11] and remanence [12], [13] methods. However, compared to the two later methods, highly sensitive measurements of magnetic susceptibility are difficult to achieve due to the interference of the excitation magnetic field. In the case of Superconducting Quantum Interference Device (SQUID) system, although the harmonics induced by the saturation characteristic of the magnetic nanoparticles can be utilized to measure their magnetic response [3], [5], [14], the large interference may destabilize the SQUID operation. Furthermore, information on the fundamental component cannot be utilized during analyzing the magnetic properties.

With the aim to achieve a high-sensitivity, low-running cost and compact evaluation tool for magnetic nanoparticles, we have developed an integrated AC/DC magnetometer using high-T_c SQUID. As the developed system is equipped with DC and AC magnetization measurement functions, two different coil configurations were adopted previously for both functions, respectively, to reduce the interference in the AC magnetization measurement. However, these two detection coil configurations had resulted in a complex detection unit and spatial limitation at the sample circumference. Moreover, simultaneous measurements of the AC and DC magnetizations are difficult and may require two SQUIDs to operate at a same time. To achieve a compact detection unit as well as reducing the interference of the excitation magnetic field in our developed system, we proposed a compensation coil technique during the fabrication of the detection coil. In comparison to the sensitive axis alignment technique used in the previous system, the compensation coil technique reduced the interference effectively. Moreover, a compact detection unit was achieved, operable using one SQUID for both functions.

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II. Experimental

A. Measurement System

The developed system is shown in Fig. 1. A flux transformer was used, consists of normal Cu coils as a detection coil and a superconducting input coil which is inductively coupled to a high-T_c SQUID. The ramp-edge Josephson junctions of the high-T_c SQUID was fabricated using advanced multilayer fabrication technique by ISTEC, Japan. In the DC function, a DC magnetic field is applied to a sample and its magnetization signal is modulated by vibrating it perpendicularly to the sensitive axis of the detection coil. Then, the magnetization signal is lock-in detected. In the AC function, the dynamic magnetization of a stationary sample is measured during application of AC and DC magnetic fields. A secondary applied coil was used in a computer-controlled electromagnet to improve the magnetic field resolution.

B. Detection Coil Optimization

In the previous system, a planar differential coil was used to reduce the environmental noise for the DC function. Moreover, we have optimized the geometry of the detection coil to improve the sensitivity by using elliptical coils [15]. The sensitive axis of the planar differential coil was placed parallel to the excitation magnetic field as shown in Fig 1. However, due to the large interference of the dynamic AC magnetic field, the sensitive axis was placed perpendicularly to the excitation magnetic field. An axial differential coil was used for the AC function, resulting in two detection coils in a system.

Although the residual signal due to the excitation magnetic field in the AC function was reduced to some extent, this had resulted in a complex detection unit. Moreover, substantial reduction of the residue signal was difficult and technically

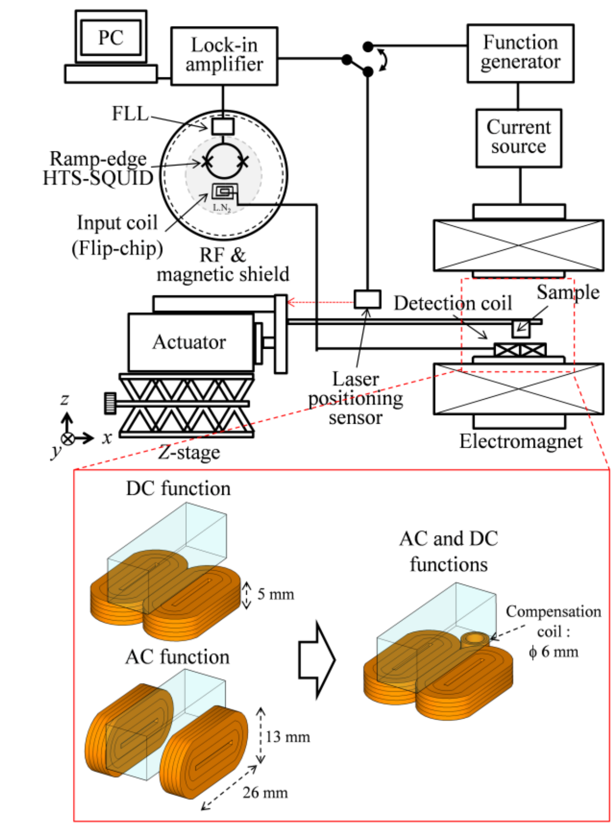

Fig. 1. Schematic diagram of the developed system and coil arrangements for AC and DC functions

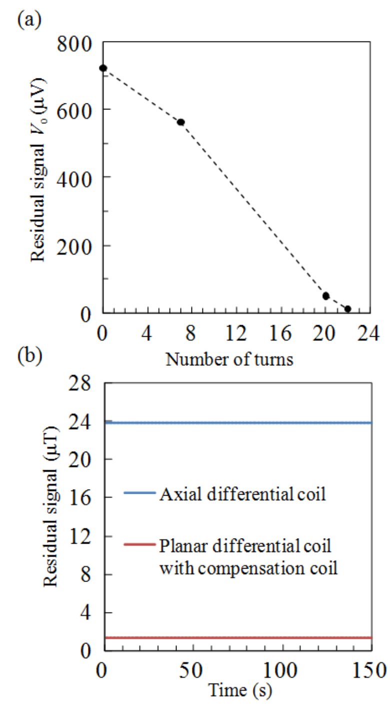

Fig. 2. (a) Induced voltage of the planar differential coil with respect to the number of turns of the compensation coil. (b) Residual magnetic signal in the compensation coil and previous sensitive axis alignment techniques at 20 mT_{pp} of 5 Hz AC magnetic field.

challenging. This was due to the slight difference in the intrinsic characteristics and alignments of the elliptical coils, respectively, despite of they were fabricated identically. A more robust and simple method is desired. The cancelling of the interference can be performed using mechanical and electronic methods [16]. In the mechanical method, as same in the previous system, the sensitive axis can be aligned to obtain higher cancelling rate. However, this will interfere with the spatial alignment of the sample circumference. In the electronic method, the interference of the excitation magnetic field can be regarded as an in-phase noise. By adding an in-phase signal electronically or magnetically to the detection system, the interference of the excitation magnetic field can be cancelled. However, this method is difficult and resulting to a complex detection circuit. As the elliptical coils were identically fabricated, only small compensation signal is required. Furthermore, the in-phase compensation signal can be supplied by the excitation magnetic field. For these reasons, we used a small compensation coil in series with the planar differential coil as shown in Fig. 1. We tuned the compensation coil manually to optimize the cancelation rate.

III. Experimental Results

The induced voltage V_o of the planar differential and compensation coils with respect to the number of turns of the compensation coil is plotted in Fig. 2 (a). The frequency and peak-to-peak value of the AC magnetic field were 5 Hz, and 80 mT_{pp}. The induce voltage V_o was lock-in detected. The residue signal was reduced effectively when the number of turns was increased to the optimum value. The comparison of the residual magnetic signal in the compensation coil method with the previous method is shown in Fig. 2 (b). The magnetic residual signal was measured by connecting the detection coil to the SQUID under 20 mT_{pp} of 5 Hz AC magnetic field. It can be shown that the compensation coil method reduced greatly the magnetic residual signal and resulted in one detection coil for the AC and DC functions. Furthermore, as product of the number of turns and flux-coupling area of the compensation coil in comparison with the elliptical coil was relatively small by a factor of 300, the effect of the compensation coil during sample measurements was negligible. As the sensitive axis of the detection coil is parallel to the magnetization axis, improvement in the sensitivity can be expected.

IV. Conclusion

We have proposed and investigated the compensation coil method to be applied in the developed AC/DC High-T_c SQUID magnetometer. The detection coil combined with the optimized compensation coil reduced the interference of the excitation magnetic field effectively. The compensation coil method had resulted in a compact detection system with a single detection coil for both functions and a reduced spatial limitation for the sample shape. High-sensitive measurement of magnetic nanoparticles can be expected using the developed system.

REFERENCES

[1] K. Enpuku, A. Ohba, K. Inoue, and T. Q. Yang, "Application of HTS SQUIDs to biological immunoassays," *Phys. C Supercond.*, vol. 412–414, pp. 1473–1479, Oct. 2004.

[2] S. Y. Yang, J. J. Chieh, C. C. Yang, S. H. Liao, H. H. Chen, H. E. Horng, H. C. Yang, C. Y. Hong, M. J. Chiu, T. F. Chen, K. W. Huang, and C. C. Wu, "Clinic Applications in Assaying Ultra-Low-Concentration Bio-Markers Using HTS SQUID-Based AC Magnetosusceptometer," *IEEE Trans. Appl. Supercond.*, vol. 23, no. 3, pp. 1600604–1600604, Jun. 2013.

[3] B. Gleich and J. Weizenecker, "Tomographic imaging using the nonlinear response of magnetic particles.," *Nature*, vol. 435, no. 7046, pp. 1214–7, Jun. 2005.

[4] T. Yoshida, N. B. Othman, T. Tsubaki, J. Takamiya, and K. Enpuku, "Evaluation of Harmonic Signals for the Detection of Magnetic Nanoparticles," *IEEE Trans. Magn.*, vol. 48, no. 11, pp. 3788–3791, Nov. 2012.

[5] J. B. Weaver, A. M. Rauwerdink, C. R. Sullivan, and I. Baker, "Frequency distribution of the nanoparticle magnetization in the presence of a static as well as a harmonic magnetic field," *Med. Phys.*, vol. 35, no. 5, p. 1988, 2008.

[6] N. L. Adolphi, D. L. Huber, J. E. Jaetao, H. C. Bryant, D. M. Lovato, D. L. Fegan, E. L. Venturini, T. C. Monson, T. E. Tessier, H. J. Hathaway, C. Bergemann, R. S. Larson, and E. R. Flynn, "Characterization of magnetite nanoparticles for SQUID-relaxometry and magnetic needle biopsy.," *J. Magn. Magn. Mater.*, vol. 321, no. 10, pp. 1459–1464, May 2009.

[7] C.-Y. Hong, C. C. Wu, Y. C. Chiu, S. Y. Yang, H. E. Horng, and H. C. Yang, "Magnetic susceptibility reduction method for magnetically labeled immunoassay," *Appl. Phys. Lett.*, vol. 88, no. 21, p. 212512, 2006.

[8] K. Enpuku, T. Tanaka, Y. Tamai, and M. Matsuo, "AC susceptibility of magnetic markers in suspension for liquid phase immunoassay," *J. Magn. Magn. Mater.*, vol. 321, no. 10, pp. 1621–1624, May 2009.

[9] S.-H. Chung, A. Hoffmann, K. Guslienko, S. D. Bader, C. Liu, B. Kay, L. Makowski, and L. Chen, "Biological sensing with magnetic nanoparticles using Brownian relaxation (invited)," *J. Appl. Phys.*, vol. 97, no. 10, p. 10R101, 2005.

[10] K. Enpuku, T. Tanaka, T. Matsuda, F. Dang, N. Enomoto, J. Hojo, K. Yoshinaga, F. Ludwig, F. Ghaffari, E. Heim, and M. Schilling, "Properties of magnetic nanoparticles in the Brownian relaxation range for liquid phase immunoassays," *J. Appl. Phys.*, vol. 102, no. 5, p. 054901, 2007.

[11] D. Eberbeck, C. Bergemann, S. Hartwig, U. Steinhoff, and L. Trahms, "Binding kinetics of magnetic nanoparticles on latex beads and yeast cells studied by magnetorelaxometry," *J. Magn. Magn. Mater.*, vol. 289, pp. 435–438, Mar. 2005.

[12] A. Tsukamoto, K. Saitoh, D. Suzuki, N. Sugita, Y. Seki, A. Kandori, K. Tsukada, Y. Sugiura, S. Hamaoka, H. Kuma, N. Hamasaki, and K. Enpuku, "Development of Multisample Biological Immunoassay System Using HTS SQUID and Magnetic Nanoparticles," *IEEE Trans. Appl. Supercond.*, vol. 15, no. 2, pp. 656–659, Jun. 2005.

[13] K. Enpuku, K. Soejima, T. Nishimoto, H. Tokumitsu, H. Kuma, N. Hamasaki, and K. Yoshinaga, "Liquid phase immunoassay utilizing magnetic marker and high T[sub c] superconducting quantum interference device," *J. Appl. Phys.*, vol. 100, no. 5, p. 054701, 2006.

[14] T. Yoshida, K. Ogawa, T. Tsubaki, N. B. Othman, and K. Enpuku, "Detection of Magnetic Nanoparticles Using the Second-Harmonic Signal," *IEEE Trans. Magn.*, vol. 47, no. 10, pp. 2863–2866, Oct. 2011.

[15] M. M. Saari, K. Sakai, T. Kiwa, and K. Tsukada, "Optimization of the Detection Technique for a Vibrating-Sample Magnetometer Using High-Tc SQUID," *IEEE Trans. Appl. Supercond.*, vol. 23, no. 3, pp. 1600204–1600204, Jun. 2013.

[16] K. Tsukada, T. Kiwa, and H. Tahara, "Highly Sensitive Measurement of Moisture Content Using HTS-SQUID," *IEEE Trans. Appl. Supercond.*, vol. 19, no. 3, pp. 878–881, 2009.