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https://doi.org/10.15017/1516048

出版情報：九州大学大学院システム情報科学紀要. 9 (2), pp.61-65, 2004-09-24. 九州大学大学院システム情報科学研究院
バージョン：
権利関係：

九州大学大学院
システム情報科学紀要
第9巻 第2号 平成16年9月

Research Reports on Information Science and
Electrical Engineering of Kyushu University
Vol.9, No.2, September 2004

Detection of Magnetic Nanoparticles Utilizing AC Susceptibility Method with Normal Pickup Coil

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(Received June 8, 2004)

Abstract: We developed a system to detect the magnetic nanoparticles utilizing AC susceptibility measurement. In this system, room temperature pickup coil made of copper wire was used as a sensor to detect the magnetic signal from the particles. In this method, an AC excitation field was applied to the particles, and the resulting signal field from the particles was detected with pickup coil. We used the differential double pickup coils with special configuration in order to reduce the coupling of the excitation field to the pickup coil and to suppress an additional magnetic noise from a substrate. With this configuration, the coupling of the excitation field to the pickup coil can be reduced to less than $1/10^6$, and the additional noise from substrate can be also eliminated. We also increased the number of turns of pickup coil and used the sample with high magnetic susceptibility in order to enhance the detectable field signal from particles. As a result, the minimum detectable weight of particles reaches to 1 ng. With experimental result and simulation, we calculated the magnetic susceptibility of particles in applied excitation field.

Keywords: Magnetic nanoparticles, Pickup coil, AC susceptibility, Biological immunoassay, SQUID

1. Introduction

Recently, magnetic nanoparticles have been applied to biological immunoassay, i.e., detection of the binding reaction between an antigen and its antibody [1),2)] In this application, the antibody is labeled with the magnetic nanoparticles, and the binding reaction is detected by measuring the magnetic field from the nanoparticles. Several sensors have so far been developed for this application, such as Hall probes [3)], giant magneto-resistance arrays [4)], atomic force microscopy [5)], and superconducting quantum interference devices (SQUIDs) [1),2),6),7)].

Due to the advantages of easy handle and low cost, a room temperature normal pickup coil made of copper wire was used to detect the magnetic field from nanoparticles [8)]. The room temperature normal pickup coil connected with SQUID was also used in magnetometer [9)], and successfully applied to the measurement of impedance- magnetocardiogram (I-MCG) signals [10)].

For the detection of the magnetic nanoparticles, three methods have so far been developed, i.e., susceptibility, relaxation and remanence measurements. **Table 1** shows the differences of three methods in the detection of magnetic nanoparticles. Among these methods, susceptibility measurement has an advantage that the magnetic signal is determined only by the total weight of the particles and is almost independent of the size of the particles. On the other hand, the magnetic signal depends strongly on the size of the particles in the case of relaxation and remanence measurements.

In the susceptibility measurement, however, the magnetic signal must be measured in the presence of the excitation field. This excitation field is much larger than the magnetic signal from the particles. Therefore, key point in this measurement method is to minimize the interference from the excitation field [1),2),7)].

In this paper, we present a detection of magnetic nanoparticles with AC susceptibility measurement. In this method, pickup coils made of copper wire with special configuration are used as a sensor to detect the signal from nanoparticles. In the experiment, an AC excitation field is used to magnetize the particles, and the resulting signal field from the particles is detected with pickup coil. First, we describe the configuration of coils. The performance of system is improved by using differential double pickup coils that can cancel the additional field noise from substrate. The coupling of the excitation field to the pickup coil can be reduced to less than $1/10^6$ by using mechanical and electrical adjustments. Next, using this system, we show the detection of the Fe_3O_4 particles. The minimum de-

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Table 1 Methods to detect magnetic nanoparticles.

Methods	Excitation field B_{ex}	Advantage	Disadvantage
Susceptibility	DC or AC field (1 mT)	any particles	detection inside B_{ex}
Relaxation	pulse field (1 mT)	detection outside B_{ex}	strong size effcet
Remanence	outside the system (0.1 T)	detection outside B_{ex}	strong size effect

tectable weight of particles reached to 1 ng. Finally, using the experimental value, we calculate the magnetic susceptibility of the particles.

2. Pickup Coil Design and Improvement

In previous study [8], we developed a measurement system for the detection of the magnetic nanoparticles utilizing AC susceptibility method. Using this system, we can detect Fe_3O_4 magnetic nanoparticles down to the weight of 7 ng at room temperature. However, improvement of the system performance is desired since the minimum detectable value was limited by the additional noise from the substrate.

In the measurement, the substrates used for nanoparticles are usually made of glass or plastic. When a larger external field is applied, these substrates show magnetization due to the impureness of substrate. The additional magnetic field noise produced by the substrates is comparable to or larger than the magnetic signal from the nanoparticles when the weight of the particles becomes less than 7 ng. Therefore, it is desired to reduce the field noise from the substrate.

In order to improve the system performance, we adopted the differential method, i.e., double-pickup coils instead of single-pickup coil in the previous study. This double-pickup coils can cancel the additional magnetic field noise from the substrate. The configuration of coils is shown in **Fig. 1(a)**, which consisted of three coils made of copper wire, i.e., the pickup coil L_p for the detection of the magnetic signal, the field coil L_m for applying the magnetic field to the nanoparticles, and the compensation coil L_c. Here, two identical coils were used for pickup coil at lower position, which were separated apart by a distance of about 5 mm, and the direction the winding of the two coils was opposite each other. This double pickup coils were made of copper wire of 80 mm diameter, and the average radius of the coil was $r = 2.5$ mm. In the measurement, the sample passed just beneath only one of two coils. The size of sample is 3 mm diameter in an area, which is smaller than the distance of two coils. Another pickup coil was placed at upper position, and apart from the center of field coil L_m by a few millimeters. This coil was made of wire of 80 mm diameter, and number of turns was 50.

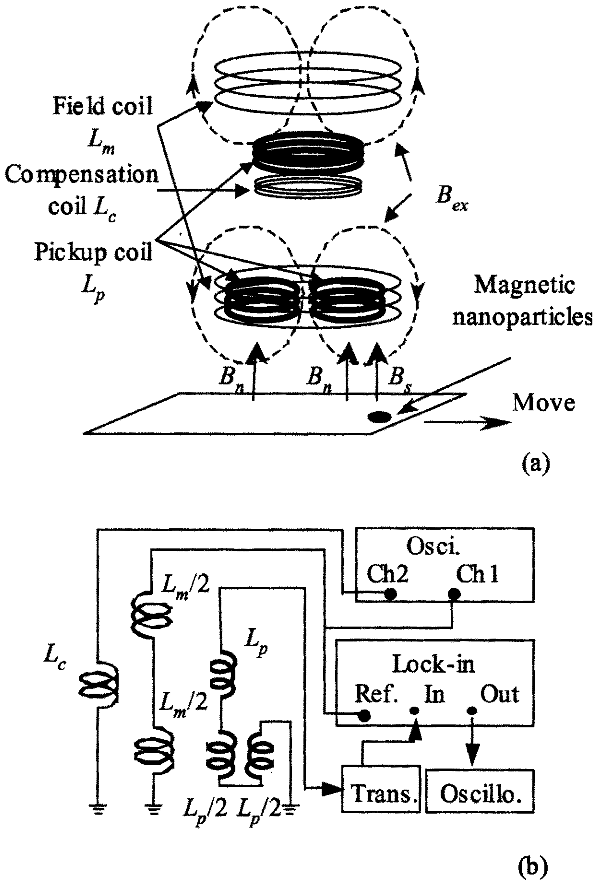

Fig. 1 AC susceptibility measurement system with normal pickup coil for the detection of the magnetic nanoparticles. (a) Configuration with differential double pickup coils. (b) Equivalent circuit of the system.

The field coil L_m consisted of two identical coils made of copper wire of 200 mm diameter, which were mounted outside of the pickup coils and separated apart by a distance of about 25 mm, but the direction of the winding of two coils was opposite. Compensation coil L_c was at the middle of pickup coil, and the number of turns was 50.

Compared to the previous system, we increased the number of turns from n= 100 to 200 for the double pickup coils, since the detected voltage signal V_s

is proportional to the number of turns of pickup coil. We also increase the excitation field B_{ex} by increasing the numbers of turns of field coil L_m from 150 to 200, since the resulting signal field B_s increase in proportion to the excitation field B_{ex}. After these improvements, the detected signal much increased.

3. AC Susceptibility Measurement

Since the signal field B_s from nanoparticles is much smaller than the excitation field B_{ex}, it is important to avoid the interference from excitation field B_{ex}. It is also desired to reduce the field noise B_n from the substrate. As shown in **Fig. 1(a)**, the field $B_{ex} + B_n + B_s$ couples to one of double coils, while the filed $-(B_{ex} + B_n)$ couples to another of double coils. In the ideal case, the field $(B_{ex} + B_n)$ are completely canceled by the differential double pickup coils, and only the signal field B_s is detected, i.e., the additional interference B_n from the substrate is eliminated.

However, due to the larger value of the excitation field B_{ex} and small asymmetry of double coils, the excitation field B_{ex} can not be completely canceled at double pickup coils. Therefore, we first carefully adjusted the position of the double coils relative to the center of the field coil L_m. Another coil at upper position was also set to cancel B_{ex}. With this configuration and mechanical adjustment, the coupling of the excitation field to the pickup coil can be canceled each other to about $1/10^3$ of B_{ex}. Then, we used the compensation coil L_c for further cancellation by the pickup coil at upper position. By finely tuning the amplitude and the phase of the AC current to the compensation coil, we can compensate the residual coupling in the pickup coil to less than $1/10^3$. As a result, the total coupling of the excitation field to the pickup coil can be reduced to less than $1/10^6$ of B_{ex}. Therefore, we can measure very small magnetic signal B_s of a few pT from nanoparticles.

Figure 1(b) shows the equivalent circuit of the measurement system. As shown, the oscillator supplies AC current to the field coil L_m and the compensation coil L_c by Ch1 and Ch2, respectively, where the amplitude and phase of the AC current can be carefully adjusted. The transformer was used to amplify the magnetic signal from the pickup coil L_p by a factor of 10, and the lock-in amplifier was used to detect the signal.

In the experiment, an AC current with a frequency of $f = 25$ kHz was supplied to the field coil L_m. Then, the excitation field B_{ex} of about 1 mT was applied in perpendicular to the magnetic nanoparticles, which were placed on a plastic substrate, and moved at a speed of 55 mm/s. When the nanoparticles passed just beneath the one of double pickup coils, the resulting signal field from the particles was lock-in detected. Here, the output of the lock-in Amplifier is band passed from 1 Hz to 20 Hz.

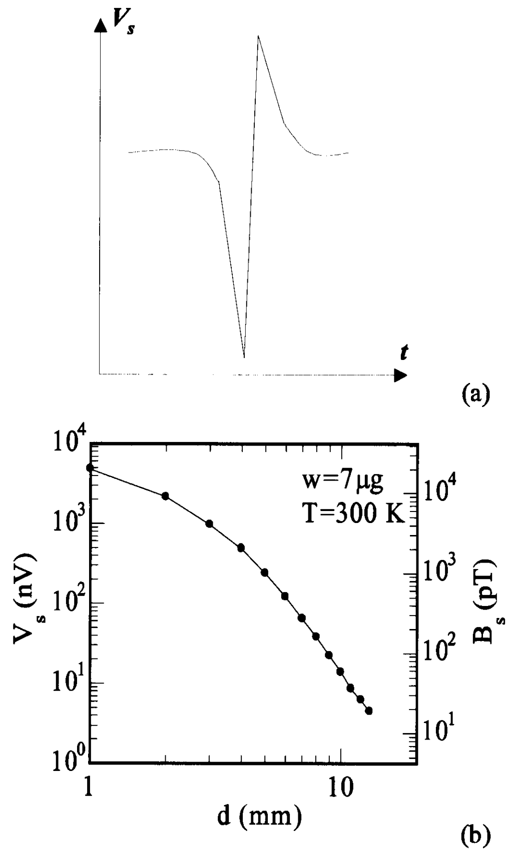

Fig. 2 (a) The typical output signal of the lock-in-amplifier when the magnetic nanoparticles were moved beneath the pickup coil. (b) Dependence of the magnetic signal on the distance d between pickup coil and the particles. The particles of of A with weight $w = 7$ μg was used in the measurement.

Two samples, A with low magnetic susceptibility and B with high magnetic susceptibility, were used in the measurement.

4. Results and Discussions

The typical waveform of the detected signal is shown in **Fig. 2(a)**, which was obtained when the particles passed beneath the pickup coil. The peak value of the signal becomes proportional to the amount of the Fe_3O_4 magnetic particles.

Figure 2(b) shows the dependence of the magnetic signal on the distance d between the pickup coil and the particles. In the experiment, the pickup coil with number of turns $n = 100$ and sample A with weight of $w = 7$ μg were used. The distance d between the pickup coil and the particles was changed from 1 mm to 10 mm. The left side of the vertical axis shows the voltage V_s across the pickup coil, while the right side shows the average magnetic filed B_s at the pickup coil. Here, the relation between V_s and B_s was given by $V_s = 2\pi f n(\pi r^2)B_s$.

As shown, the magnetic signal strongly depends on the distance d. When the distance d is close to 1 mm, the magnetic signal changed by $1/d$. However, the magnetic signal changed rapidly by $1/d^3$ when the distance becomes larger than 5 mm. Therefore, in order to increase the signal from nanoparticles, the pickup coil is expected to be near the sample as closely as possible.

We also measured the magnetic signals with different weight w of the particles. In the measurement, the distance between the pickup coil and the particles was $d =$ 1mm. Curve (b) in **Fig. 3** shows the result when sample A was used. As shown, linear relationship was obtained between the magnetic signal and the weight w of the particles. For this case, the minimum detectable weight was down to 3.5 ng. For comparison, curve (a) shows the result measured in previous study. As shown, this value of 3.5 ng in curve (b) was 2 times better than the minimum detectable weight of 7 ng in curve (a). This improvement is mainly due to the introduction of the differential double pickup coils that can cancel the additional field noise from substrate.

It is well known that the magnitude of the signal field from particles depends not only on the applied excitation field B_{ex}, but also on the magnetic susceptibility of the particles itself, i.e., the resulting signal becomes larger for the particles with higher magnetic susceptibility. Therefore, in order to detect the minimum weight of particles more efficiently, sample B with high magnetic susceptibility was used. Curve (c) shows the result. Similarly, the linear relationship was obtained, and the minimum detectable weight was down to 1 ng. This value is 3.5 times better than the curve (b), and 7 times better than the curve (a). Here, the signal voltage across the pickup coil was $V_s = 4$ nV, corresponding to the signal field of $B_s = 6.5$ pT. This result suggests that it is effective to use particles with high magnetic susceptibility.

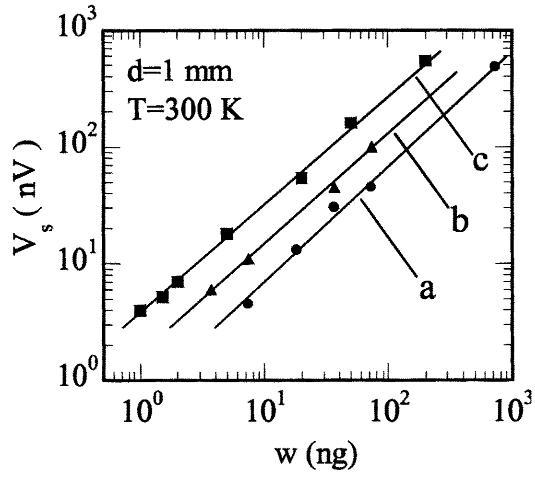

Fig. 3 Relationship between the weight of particles and the detected signal. Curve (a) shows the result of sample A with the configuration of single pickup coil of $n = 100$. Curve (b) and (c) show the result of sample A and B with the configuration of double pickup coils of $n = 200$, respectively.

From the above results, we can estimate the value of susceptibility χ of the magnetic particles with AC susceptibility method. **Fig. 4(a)** shows the arrangement of the pickup coil for the case of number of turns $n = 10 \times 10$ in measurement. As shown, the distance from sample and the diameter of each turn of pickup coil change with the sequence from position A to D.

Using the configuration of the pickup coil, we can calculate the magnetic signal detected with the pickup coil. **Fig. 4(b)** shows the simulated result of magnetic flux that interlinks to each of the pickup coil. As shown, the signal flux changes with the sequence of pickup coil from $N = 1$ to 100. For this case, therefore, the total signal flux is calculated as $\Phi_s = 0.944 \times \mu_0 m_z / 4\pi$ [Wb]. Here, the value of m_z can be expressed as $\mu_0 m_z = \mu_0 M_z \times NV/A = \mu_0 M_z \times w/A\rho$. Therefore, we obtain the following relationship

$$\Phi_s = \frac{0.944}{4\pi} \times \mu_0 M_s \times \frac{w}{A\rho} \tag{1}$$

where μ_0 is the space permeability, m_z is the magnetic moment of the sample per unit area, M_z is the average magnetization of the sample, V is the volume of each particle, and A, N w, $\rho = 5.2$ are the area, amount, weight and specific gravity of particles of Fe_3O_4, respectively.

For the curve (a) in **Fig. 3**, the applied excitation field B_{ex} is about 0.76 mT. The minimum detectable weight is 7 ng, corresponding to the signal voltage of $V_s = 4.5$ nV across the pickup coil. From the relationship $V_s = 2\pi\Phi_s f$, we can obtain the flux $\Phi_s = 2.87 \times 10^{-14}$ [Wb] from the measure-

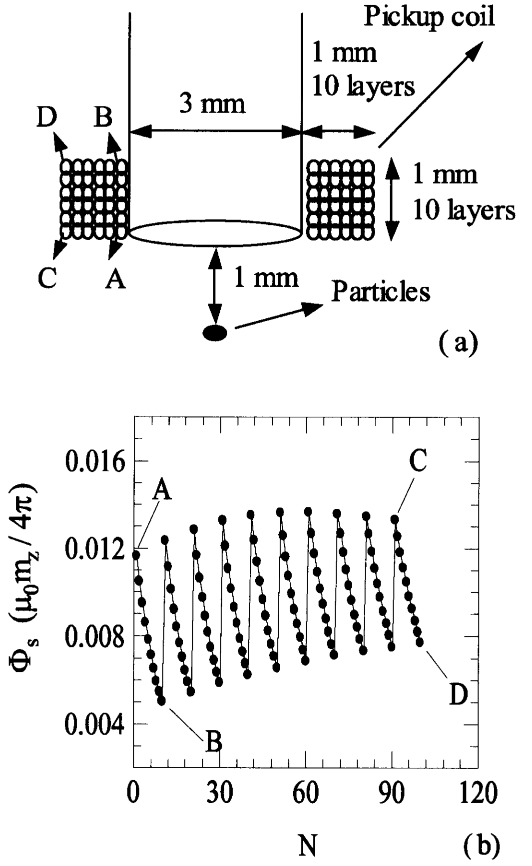

Fig. 4 (a) Arrangement of the pickup coil. The pickup coil at four different positions are indicated by A, B, C and D, and distance from sample and the size of each turn of pickup coil change by the sequence from A to D. (b) Dependence of the simulate signal flux detected by each turn of pickup coil on the sequence from $N = 1$ to 100.

ment. Substituting this value into Eq.(1), we can estimate the average magnetization as about $\mu_0 M_z = 2$ mT. Therefore, the magnetic susceptibility can be estimated as $\chi = \mu_0 M_z / B_{ex} = 2.63$ for the particles of sample A. For sample B, the magnetic susceptibility χ is estimated by about 3 times higher than the sample A.

5. Conclusions

Normal pickup coil made of copper wire was used as a sensor for the detection of the magnetic particles with AC susceptibility measurement. In this method, the differential double pickup coils with special configuration were designed to cancel the additional magnetic noise from the substrate. With this configuration, the coupling of the excitation field to the pickup coil can be reduced to less than $1/10^6$ by using mechanical and electrical adjustments. The additional noise from substrate can also be effectively eliminated. Furthermore, by increasing the number of turns of pickup coil and using the sample with high magnetic susceptibility, the detectable signal field from particles was much enhanced. As a result, the minimum detectable weight of particles reaches to 1 ng. This value is 7 times better than the previous reported value of 7 ng. By calculation, it was shown the magnetic susceptibility χ of samples A and B were about 2.63 and 7.9, respectively.

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