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Multi-Frequency ESR Using a Microcantilever in the Millimeter Wave Region

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Abstract. Low-temperature magnetic properties of condensed matter system is investigated microscopically with high-frequency electron spin resonance (ESR). Experiments are carried out conventionally with a transmission method, but its sensitivity is not often sufficient to detect ESR signals of systems with small number of spins such as newly synthesized microcrystals. To attain better sensitivity, we focus on a novel technique of multi-frequency ESR system utilizing a microcantilever. In this method, ESR signal is detected as a torque change associated with the absorption of electromagnetic wave. Its sensitivity is greatly increased due to mechanical resonance when the modulation frequency of electromagnetic wave coincides with the eigenfrequency of the cantilever. In this study, we have succeeded in mechanical detection of ESR signals in the millimeter wave region up to 240 GHz. The achieved signal-to-noise ratio was greater than $10^3$ for 1-µg sample of Co-Tutton salt, corresponding to the spin sensitivity of $10^9$ spins/G.

1. Introduction
High-frequency electron spin resonance (ESR) has unique advantages such as high spectral resolution or observation of broad resonance. In particular, this technique has been applied to investigate low-temperature magnetic properties of interesting spin systems such as quantum spin systems. Experimentally, high-frequency ESR is carried out using a transmission method [1], in which one measures the transmitted intensity of electromagnetic wave through the waveguide containing a sample. For this setup, spectroscopic measurement can be performed by changing the frequency of the light source. On the other hand, its sensitivity is inevitably worse than that of cavity perturbation technique. This indicates that large amount of sample volume (~10 mg) is needed for reliable measurement.

To attain better sensitivity in the high frequency region, we focus on a novel ESR technique, based upon mechanical detection using a microcantilever. In this method, one detects a change of magnetic torque associated with ESR absorption of the sample mounted on the cantilever. Since the cantilever size is of order of 100 µm, a piece of microcrystal can be measured with
high precision. The most remarkable feature of this technique is that electromagnetic wave is introduced into the sample position using a waveguide in a similar manner to the transmission method. Therefore, the frequency range can be extended to terahertz region with keeping high sensitivity.

Previously, we reported ESR detection using the cantilever [2], but its signal-to-noise ratio of at most ten was still insufficient for practical use. In this study, we have improved its detection scheme using a lock-in amplifier and have successfully increased the sensitivity by three orders of magnitude. This improvement leads to mechanical detection of ESR signals in the millimeter wave region up to 240 GHz.

2. Experimental setup
Figure 1 shows the cantilever used in this study. This cantilever was commercially available and its dimensions were $400 \times 50 \times 4 \, \mu m^3$ [3]. Torque acting on the cantilever was detected as a change of piezoresistance fabricated at cantilever legs. A compensation lever is also fabricated on the same substrate to eliminate the effect of magnetoresistance and thermal drift of the piezoresistance. Details of the cantilever are given in ref. [4].

The detection scheme developed in this study was as follows: First, the output of Gunn oscillator was electronically modulated using an external oscillator. The modulated electromagnetic wave was transmitted through a waveguide from the top of the cryostat, and was irradiated to a sample mounted on the cantilever. The cantilever was dc biased, so that the output voltage from the cantilever bridge was modulated when ESR absorption occurred. This is because magnetization change, and thereby torque change, was induced by the absorption of electromagnetic wave. The signal was detected using a lock-in amplifier which was synchronized to the external oscillator. This method is advantageous for the following two reasons: first, background signals can be eliminated, since the magnetization change associated with on-off operation of the electromagnetic wave is measured. Second, as described later, ESR signal can be amplified by mechanical resonance of the cantilever, when the modulation frequency coincides with the eigenfrequency of the cantilever.

The sample used in this study was a single crystal of Co-Tutton salt. The sample size was typically 50-100 $\mu m$ in length, and the mass was estimated to $\sim 1 \, \mu g$ from the change of eigenfrequency. Experiments were carried out using a 4 T superconducting magnet. The
Figure 3. ESR signal intensity versus modulation frequency. The value is normalized by the peak intensity.

Figure 4. Multi-frequency ESR in the millimeter wave region up to 240 GHz. High-frequency data are magnified for clarity.

cantilever was kept in $^4$He gas atmosphere to ensure the thermal contact. Sample temperature was slightly warmed up to $\sim$5 K during experiments, because of the electromagnetic wave irradiation.

3. Results and Discussion

Figure 2 shows typical ESR absorption lines at 5 K and 80 GHz. We clearly observed two resonance dips at 1.25 and 1.95 T, which can be attributed to the ESR signals from Co$^{2+}$ ion. The corresponding $g$ values are 4.93 and 3.16, respectively, being consistent with reported large anisotropy of $g$ value [5]. According to the crystallographic analysis, there exist two independent Co sites in the crystal [6, 7], which are magnetically coupled by weak exchange coupling of order of $J \sim 0.01$ K [8]. Considering that magnetic field is strong enough to satisfy the condition $g\mu_B B > J$ in the present case, two observed ESR lines are explained by the so-called exchange splitting.

As also shown in Figure 2, we found strong enhancement of the ESR absorption line, depending on the modulation frequency. When the intensity of electromagnetic wave is modulated at ESR absorption, the periodic change of magnetization is induced so as to excite the cantilever oscillations. This effect results in the mechanical resonance of the cantilever. Figure 3 shows the normalized intensity of the low-field absorption in Figure 2 versus modulation frequency of Gunn oscillator. Sharp increase of the ESR signal intensity is found at $f_{\text{mod}}=9200$ Hz, which exactly corresponds to the cantilever’s eigenfrequency. Since the cantilever was kept in gas atmosphere, high quality factor of $\sim$300 was attained.

We obtained the resultant signal-to-noise ratio exceeding $\sim 10^3$ for 80 GHz. With the sample mass of 1 $\mu$g, the minimum detectable spin number is estimated to $\sim 10^{12}$ spins in the present case. Considering the absorption line width of 10 mT, the detectable spin sensitivity was $\sim 10^9$ spins/G at 80 GHz, which is comparable to that of commercial X-band ESR system. This corresponds to the improvement of sensitivity by three orders of magnitude from our previous results [2].

As mentioned above, the operating frequency can be easily changed by replacing the light source at the top of the cryostat. Figure 4 shows ESR absorption lines for various operating frequencies, keeping the modulation frequency of Gunn oscillator with cantilever’s eigenfrequency. Here, we focused the low-field absorption shown in Figure 2, because of the
limited field range up to 4 T. We successfully detected ESR signals in the millimeter wave region up to 240 GHz. Thus, our system can be used in higher frequency region than other techniques based upon similar mechanical detection [9]. The difference in the absorption intensity mainly originates from the output power of Gunn oscillator. For example, a frequency doubler was used to generate 240 GHz, so that the output power was only 4% (∼1 mW) of that for fundamental frequency.

It is of interest to mention high-frequency ESR based upon quasi-optic techniques [10, 11]. These techniques also work in a similar frequency region, and have comparable sensitivity of $10^9$ spins/G. However, their extension to the high frequency region is problematic due to the frequency-fixed optical components, in contrast to our system. Therefore, the advantage of our system is the ability to further extend the frequency region, possibly to terahertz region in the future. For this aim, we are planning to combine our system with a backward wave oscillator, which covers the frequency range between 300 and 1200 GHz and has larger output power of order of 10 mW.

4. Summary
We have developed high-sensitive ESR technique based upon mechanical detection, enabling both high-frequency and multi-frequency ESR. The achieved sensitivity was ∼$10^9$ spins/G at 80 GHz, and the frequency region was successfully extended to the millimeter wave region. The developed ESR technique would be useful to study low-temperature magnetic behavior of newly synthesized magnets, whose sample size is less than 100 µm.

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