Cantilever-detected high-frequency ESR measurement using a backward travelling wave oscillator

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Abstract. Our cantilever-detected electron spin resonance (ESR) technique is motivated for terahertz ESR spectroscopy of a tiny single crystal at low temperature. In this technique, ESR signal is detected as deflection of a sample-mounted cantilever, which is sensitively detected by built-in piezoresistors. So far, ESR detection at 315 GHz was succeeded using Gunn oscillator. In this study, we combine our ESR technique with a backward traveling wave oscillator (BWO), which can cover a wide frequency range 120-1200 GHz, to achieve better spectral resolution. Experiments were carried out at 4.2 K for a single crystal of Co Tutton salt with a newly constructed optical system. We successfully observed two ESR absorption lines in BWO frequencies up to 370 GHz. From multi-frequency measurements, the observed ESR lines shifted linearly with BWO frequency, being consistent with paramagnetic resonance. The estimated $g$ values are $g_1=3.00$ and $g_2=3.21$. The spin sensitivity was estimated to $\sim 10^{12}$ spins/gauss at 370 GHz.

1. Introduction

High-frequency electron spin resonance (ESR) is a powerful tool in the study of magnetism. High-frequency ESR technique gives a detailed insight into the local electronic structure of unpaired electrons such as transition metal ion from the analysis of the observed $g$ factor and the line width. Recently we developed a new high-frequency ESR technique using a microcantilever [1-4]. In this technique, a small magnetization change associated with ESR transition is detected as deflection of a sample-mounted cantilever. Compared to conventional transmission technique, spin sensitivity is three orders of magnitude improved. So far we achieved ESR detection at 315 GHz using Gunn oscillator [5]. However, in order to achieve better spectral resolution, operation in higher frequency region or terahertz region is strongly desired. In this study, we successfully extended the frequency region to 370 GHz using a backward travelling wave oscillator (BWO) as a light source.

In the previous report [2], we used an optical modulation technique, in which the intensity of an electromagnetic wave was modulated periodically, and its synchronized magnetization change was detected using a lock-in amplifier. We used Gunn oscillators as a light source, but its upper limit was 315 GHz, where the output power is decreased to 1 mW. In order to extend the frequency range, BWO...
Experimental setup of cantilever-detected ESR using BWO is suitable for this purpose, since it is able to cover a wide frequency range of up to 1200 GHz and we also expect relatively large output power even beyond 300 GHz. In addition, BWO is a quasi-continuous light source and suitable for spectroscopic studies to determine important parameters of spin Hamiltonian.

2. Experimental setup

In figure 1, our experimental setup used in this study is shown. Basic construction is similar to the previous system [2], but BWO is used as a light source, instead of Gunn oscillator. As a result, modulation method is changed from the previous setup. For Gunn oscillator case, modulation was done by electrically switching on and off of the power supply of Gunn oscillator. On the other hand, a mechanical chopper is used to modulate the light intensity for BWO case. The maximum modulation frequency was 1000 Hz, which is limited by the revolution speed and the slit width of the chopper blade. Namely, the slit width has to be large enough compared to the cutoff length of the electromagnetic wave. The revolution speed is controlled by an external local oscillator, and the synchronized TTL signal was fed into a lock-in amplifier as a reference signal.

BWO is a quasi-continuous terahertz light source with a relatively large output power on the order of 10 mW. By adjusting a cathode voltage, quasi-continuous electromagnetic wave can be generated. In this study, we used a BWO tube OV-30, which covers a frequency range 200-380 GHz. Since a permanent magnet is used inside the BWO, BWO was placed 1.5 m away from a 15 T superconducting magnet. The electromagnetic wave generated by BWO horizontally propagates in a copper light pipe with an inner diameter of 15 mm. Subsequently, it is reflected by a copper mirror at the top of a cryostat, and introduced into the cryostat using a stainless-steel light pipe. At the bottom of the light pipe, a brass horn was used to focus the electromagnetic wave onto a sample-mounted cantilever.

As in the previous report, we used a commercial piezoresistive microcantilever PRC400 [6]. A single crystal of Co Tutton salt was glued on a cantilever using a manipulator. Typical sample mass was 1-10 microgram. The cantilever was kept in helium gas atmosphere (P ~ 460 Torr) in order to suppress spurious cantilever vibration due to light modulation. We also used Gunn oscillators to measure ESR signal at the low frequency side.
3. Results and discussion

Figures 2 shows typical results of the multi-frequency cantilever ESR measurement using BWO. In this figure, background signals are subtracted for clarity. The background signal mainly results from imperfect compensation of the bridge circuit, even though piezoresistors are fabricated in the same conditions. The modulation frequency was 100 Hz, and the sample temperature was 4.2 K. Sample heating by irradiation was found to be negligible. Thus, experiments at lower temperatures will be possible. We clearly observed two ESR absorptions which are expected for single-crystal Co Tutton salt. As expected, the resonance field shifted to high field region with applied BWO frequency. The highest frequency was 369.7 GHz, which is a record of mechanically detected ESR measurement to our knowledge. Sample mass determined from the eigenfrequency shift was 1.1 microgram. Since the output power of OV-30 was at most 1 mW in this frequency range, the signal-to-noise ratio was limited to ten. Together with the signal-to-noise ratio, we estimated spin sensitivity on the order of $10^{12}$ spins/ gauss at 369.7 GHz.

Shown in figure 3 is a magnetic field-frequency diagram derived from figure 2, together with results obtained using Gunn oscillators. As expected for paramagnetic samples, each resonant field is well fitted by a straight line. The slope of the linear fitting gives the corresponding g factor. In the present case, fitted g factors are 3.00 and 3.21 respectively, which are consistent with those previously reported for Co Tutton salt. It is noted that the separation between two absorptions becomes wider and wider with increasing the frequency. Namely, it is about 0.2 T at 80 GHz, and is greater than 1 T for 370 GHz. The ESR line width of magnetic samples often exceeds 1 T due to strong magnetic interactions. In this sense, ESR measurement beyond 300 GHz is important.  

4. Summary

We successfully extended the frequency region of our cantilever-detected ESR technique to 370 GHz using BWO. Taking advantage of a quasi-continuous light source, multi-frequency ESR measurement beyond 300 GHz has been demonstrated. By exchanging vacuum tubes inside the BWO,
Figure 3. BWO frequency versus resonance field of Co Tutton salt. The data taken by BWO and Gunn oscillator are plotted together. Straight lines show fitted results to the data, from which the corresponding \( g \) factors are derived.

it is able to generate higher frequency (up to 1200 GHz) without changing other optical setups. Therefore, further extension toward 500 GHz is now in progress.

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