Present status and future plan of research in high magnetic fields at KYOKUGEN in Osaka University

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Abstract. After brief introduction of history and facility of the High Magnetic Field Laboratory at KYOKUGEN in Osaka University, we describe our high field and multi-frequency electron spin resonance (ESR) apparatus by utilizing pulsed and superconducting magnets for the fields up to about 60 T. For the ESR measurements in pulsed magnetic fields, several Gunn and backward oscillators, and a far infrared laser are used as millimeter and submillimeter wave sources. In steady magnetic fields up to 16 T, we have utilized a vector network analyzer with extensions which covers the frequencies between 8 and 700 GHz almost continuously. The latter ESR apparatus is used not only dense magnetic materials but also weak ones, such as metalloproteins. Therefore, we have developed high sensitive multi-frequency ESR apparatus. To extend ESR studies further, we are now constructing ESR apparatus for much higher fields up to 70 T and the wide frequency range up to 7 THz. Magnetization and transport measurements are also performed in high magnetic fields up to 70 T and 60 T, respectively and magnetization measurements under high pressure up to 1 GPa. We plan to develop a 50 T wide bore pulsed magnet with the diameter of about 50 mm for the use of high sensitive ESR and high pressure measurements above 1 GPa.

1. Introduction

The High Magnetic Field Laboratory at Osaka University headed by Date [1, 2] was founded in 1975 as the high magnetic field facility of the Faculty of Science [3]. The requirement of high magnetic field generation resulted from antiferromagnetic resonance (AFMR) experiment [2]. Non-destructive high field pulsed magnets which consist of maraging steel polyhelix coils were developed and the generation of magnetic field up to 107 T was reported [2]. In 1987, the laboratory was reorganized as a section of the Research Center for Extreme Materials (abbreviated to KYOKUGEN) of Osaka University for the development of cooperative studies with other sections [4]. Then, the center was reorganized again in 1996 including the low temperature section and was named the Research Center for Materials Science at Extreme Conditions (KYOKUGEN). The High Magnetic Field Laboratory at new KYOKUGEN was headed by the last author, Kindo and he developed a newly designed non-destructive pulsed magnet [5]. The magnet was made by winding with Cu-Ag wire and reinforced by a maraging steel cylinder and generated 80.3 T in a 10 mm bore with 7 msec pulse duration. This pulse duration is much longer than that of the previous magnet (about 0.4 msec) and is very useful for measurements of metal materials. Kindo and his coworkers developed the apparatus for magnetization and magnetoresistance measurements down to 80 mK in magnetic fields up to 60
From April of this year (2006), the center started as a new organization and is named the Center for Quantum Science and Technology under Extreme Conditions (KYOKUGEN). In the High Magnetic Field Laboratory at KYOKUGEN, we have two high magnetic field facilities. The first facility is a new one built in 1988 and the second facility is an old one built in 1975. The first and second facilities are equipped with 1.5 MJ (20 kV, 7.5 mF) and 1 MJ (13.3 kV, 11.6 mF) capacitor bank systems, respectively. The former bank system uses pressurized air gap switches and the latter one uses thyristor switches. Figures 1(a) and (b) show the pulse shapes of the magnetic field generated with pulsed magnets used at the first and second high field facilities, respectively. The pulse shape at the first facility has a short duration of about 7 msec due to the small inductance of the magnet 0.7 mH, and that at the second one has a long tail with the pulse duration of about 40 msec due to the inductance of about 7 mH. Therefore, we usually carry out magnetization and ESR measurements on high resistive samples, e.g. insulators and semiconductors at the first facility, and magnetization and transport measurements on conductive samples at the second facility. In the first facility, we also use a magnet with very short duration of about 0.4 msec made from maraging steel.

2. High field and multi-frequency ESR
As mentioned in the introduction, the High Magnetic Field Laboratory at Osaka University originated from the requirement of high magnetic field generation in AFMR measurements. In the 1960s, electron spin resonance (ESR) measurements were done by utilizing millimeter wave sources like Klystron and pulsed magnets up to about 20 T. Some remarkable findings such as spin cluster resonance in CoCl$_2$·2H$_2$O [6] were reported during this period. In the 70s and 80s, high frequency ESR measurements using HCN and H$_2$O FIR lasers were performed with short pulsed magnets made from maraging steel, but ESR experiments for the millimeter wave region were not performed because of the destruction of the Cu wave guide or resonator due to a very short pulse duration. In the late 80s, Cu-Cr-Zr-wire wound long pulsed magnets were developed and millimeter ESR measurements restarted in magnetic fields up to about 40 T. ESR spectra from the transitions within the excited triplet state in the $S=1$ one-dimensional Heisenberg antiferromagnet, the so-called Haldane magnet, were observed with this pulsed field ESR apparatus [7].

Kindo developed pulsed magnets made by winding Cu-Ag wire and reinforced by a maraging steel cylinder in the late 90s [5]. The second author, Kimura, started to build high field ESR apparatus consisting of this magnet, several Gunn oscillators, a FIR laser and an InSb
detector from 1999 [8]. This apparatus mainly uses for magnetic materials which show some field induced phase transitions at high magnetic fields [8]. Since October in 2004, the first author, Hagiwara, has transferred from the Institute of Physical and Chemical Research (RIKEN) to the High Magnetic Field Laboratory at Osaka University as the successor to Kindo bringing his multi-frequency ESR apparatus developed at RIKEN. This multi-frequency ESR apparatus is composed of a 16 T superconducting magnet (Oxford Instruments, UK) and a vector network analyzer MVNA with some extensions (ABmm, France) which covers the frequencies between 8 and 700 GHz, almost continuously. He also brought several Gunn oscillators and backward oscillators (200 and 300 GHz). Two dark gray areas below 60 T and 2000 GHz in figure 2 indicates the frequency-field windows which are covered with our present ESR apparatus.

![Figure 2. Frequency-field range covered with high field and multi-frequency ESR apparatus at KYOKUGEN in Osaka University including the range with that under construction. Inset: Multi-layer pulsed magnet made by winding Cu-Ag wire reinforced by a strong tensile textile, Zylon and a maraging steel cylinder.](image)

3. High sensitive multi-frequency ESR
High sensitive multi-frequency ESR apparatus equipped with the superconducting magnet has been developed [9] to study metalloproteins and magnetic granular thin films by the third author, Yashiro and Hagiwara since 2001 when they worked at RIKEN. To cover wide frequency range (35-600 GHz), we have chosen two types of resonators, one of which is a Fabry-Perot resonator (FPR) for the use of higher frequencies (50-600 GHz) that has the highest finess value reported so far. The others are ordinary cylindrical resonators with TE_{011} mode for 35, 50, 70, 95 and 130 GHz, because they have theoretically one order of magnitude higher sensitivity than that of the FPR. Moreover, for the latter resonators, we have achieved very stable matching under a large magnetic field sweep (e.g. 16 T) by changing the relative angle between the longitudinal axis of the wave guide and that of the cavity. By their high sensitivity and stability, we successfully observed multi-frequency ESR spectra of a metalloprotein with an integer spin system for the first time [9].

4. Magnetization and transport measurements
Magnetization measurements have been done in magnetic fields up to about 70 T at the first facility and up to about 60 T at the second facility. Transport measurements such as
magnetoresistance and Hall resistance are carried out in magnetic fields up to about 60 T at the second facility. The fourth author, Yoshii, and his collaborators have performed magnetization and magnetoresistance measurements on several kinds of rare-earth compounds, one of which is a rare-earth tetraboride RB$_4$ that possesses a unique topology characterized by orthogonal dimers equivalent to the Shastry-Sutherland lattice [10]. He has re-developed magnetization measurement apparatus under high pressure up to about 1 GPa by referring to the previous development at KYOKUGEN [5]. For precise transport measurements in steady low fields, he has also constructed similar apparatus equipped with a 12 T superconducting magnet to that used in pulsed magnetic fields.

5. Future plan
For high field multi-frequency ESR measurements, we are now developing a 70 T ESR apparatus by using a multi-layer Cu-Ag pulsed magnet as indicated in the inset of figure 2 which is used for magnetization measurements in magnetic fields up to about 70 T. To detect ESR signals at higher frequencies above 2000 GHz, we will utilize a semiconductor chip of Ge:Sb which covers the frequencies up to about 7000 GHz. The frequency-field window covered with this ESR apparatus under construction is shown by the light gray area in figure 2. As for high sensitive ESR apparatus, we will make TE$_{011}$ single mode resonators for 170 and 220 GHz and a highly stable FPR with a stable matching mechanism. In addition to these developments, construction of low temperature ESR equipments such as a 60 T pulsed field ESR apparatus down to about 0.5 K and a 16 T steady field ESR one down to about 0.1 K is now in progress.

We plan to develop a wide bore pulsed magnet with the diameter of about 50 mm which is expected to generate the magnetic fields up to about 50 T. By utilizing this magnet, it will be possible to make high field ESR apparatus with high sensitivity by introducing the ESR resonators into this magnet. In addition, we want to expand the pressure region in high magnetic field measurements up to about 50 T to obtain a wide field-pressure-temperature phase diagram for magnetic materials. Therefore, we will put high pressure cells such as an indenter-type high pressure cell into the wide bore pulsed magnet.

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