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Development of high-field and high-pressure ESR system and application to triangular antiferromagnet $CsCuCl_3$

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Abstract We have developed a new hybrid-type pressure cell for the high-pressure and high-field ESR measurement using a widely used Oxford 15 T superconducting magnet with the variable temperature insert (VTI). The size of the pressure cell was optimized and a probe was also specially designed so as to be fitted to the VTI. We confirmed that the new pressure cell can generate the pressure up to at least 2 GPa repeatedly. Using this new ESR system, high-pressure and high-field ESR measurement was performed on the triangular antiferromagnet $CsCuCl_3$ for $H \parallel c$ at 4.2 K in the frequency region 60 GHz to 420 GHz. We succeeded in observing the significant pressure effect of this compound.

Keywords High pressure ESR · Hybrid type pressure cell · $CsCuCl_3$

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1 Introduction

Pressure is one of the important parameters in condensed matter physics because it can change the interactions between electron spins continuously. Recently, novel pressure-induced phases have attracted much attention and been studied intensively especially in quantum magnets. So far we have developed high-pressure and high-field ESR systems and applied them to quantum magnets [1–6]. We use a transmission technique in our ESR systems and we have developed specially designed piston-cylinder type pressure cells using ceramics inner parts which have good transmittance to millimeter and submillimeter waves. It turned out that the high-pressure and high-field ESR measurement is a good means to explore the pressure-induced phase transition of quantum magnet like $SrCu_2(BO_3)_2$ [7].

In the latest pressure cell, whose maximum pressure is 2.5 GPa, all inner parts of the pressure cell are made with zirconium oxide and it covers the frequency region from 50 to 700 GHz. It is combined with a cryogen-free superconducting magnet with the maximum field of 10 T [4]. We use a homemade InSb hot electron bolometer as the detector. As the InSb detector cannot be used in a high field, we use a special cryostat which has enough space below the center of magnet and put it apart from the center downward. In order to use our pressure cell in a general cryostat without sufficient space below the center of the magnet, we have developed a new system in which electromagnetic wave is reflected by the mirrors at the bottom of the probe as shown in Fig. 1 (a). Outside the probe, we can use a high sensitive commercial InSb detector with its own cryostat. Although this propagation way of the electromagnetic wave is more lossy as compared with the previous 10 T system with no reflection point from top to the detector [4], we confirmed that the sensitivity is unchanged or more improved by the use of the commercial InSb detector. Using this technique, we succeeded in performing ESR measurement with the world highest cryogen-free superconducting magnet up to 25 T in the user facility of Tohoku University [6].

In this study, we apply this technique to the widely used Oxford superconducting magnet with the variable temperature insert (VTI). As the bore of the magnet is 52 mm as usual and the diameter of the VTI is 37 mm, we have to adjust the size of the pressure cell. We show the new optimized pressure cell for this standard magnet and application result of a quantum spin system $CsCuCl_3$.

2 Outline of New ESR System

We employ a superconducting magnet with the maximum magnetic field of 15 T (Oxford Instruments plc). Figure 1 (a) shows the schematic view of the new system. The inner diameter of the VTI is 37 mm and the height from the bottom to the center of the magnet is 55 mm as shown in Fig. 1 (b), where the newly developed pressure cell is shown in Fig. 1 (c). The previous pressure cell [4] is shown in Fig. 1 (d) for comparison. For the previous pressure cell, its outer and inner diameters of the cylinder are 28 mm and 5 mm, respectively, and the height from the bottom to the magnet center is 31 mm. For the new pressure cell, the outer and inner diameters of the cylinder are 23.5 mm and 5 mm, respectively, and the height from the bottom to the magnet center is 21.5 mm. The hybrid-type cylinder consisting of an inner NiCrAl cylinder and an outer CuBe sleeve is also employed.

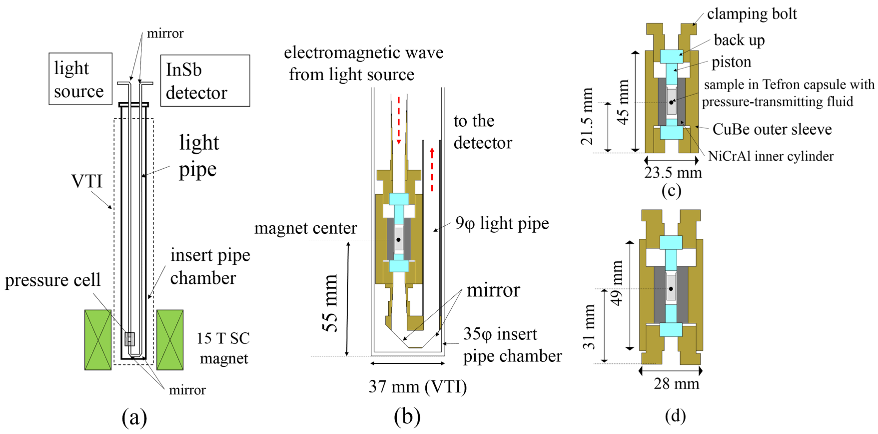

Fig. 1 (a) Schematic view of new ESR system, (b) lower part of new ESR system with VTI, (c) newly designed smaller pressure cell and (d) previous pressure cell [4].

In general, the reduction of the inner and outer diameter ratio of the cylinder reduces its strength. However, the design of the previous cell [4] was originally developed for the maximum pressure of 4 GPa [8] while the maximum pressure in our cell is limited by the toughness of the ceramics. Therefore, its design can be optimized to match the upper limit of the toughness of the ceramics. The light pipe of 9ϕ and the same sample space ($5\phi\times\sim 10$ mm) are required to keep the sensitivity [6], while the whole probe is inserted into an insert pipe chamber with the outer diameter of 35 mm with the He exchange gas [Fig. 1 (a)]. Therefore, we decided to set the outer diameter of the cylinder as 23.5 mm for the new pressure cell. The bottom clumping bolt is made like a set screw and is implanted in the body to reduce the height from the bottom to the magnet center (21.5 mm). Top and bottom clamping bolts are connected to the light pipes by the horns. The taper of the top horn is 2°, while that of the bottom horn is 3° to reduce its length. Thus, the whole probe including the new pressure cell can be fitted to the VTI.

Next, we performed the pressure generation tests for the new pressure cell. We measured the superconducting transition temperature T_c of tin by the AC magnetic susceptibility measurement to determine the pressure at low temperature [9]. Fig. 2 (a) shows the temperature dependence of the AC susceptibility obtained by several loads at room temperature. As shown in Fig. 2 (a), the abrupt decrease of the susceptibility, which shows the superconducting transition, is clearly observed and the transition temperature decreases as the load is increased. The pressure is evaluated from the relationship between the change of the transition temperature from that at ambient pressure ΔT_c and the pressure P [10]. The maximum pressure of this experiment is obtained to be 2.06 GPa. Fig. 2 (b) shows the relationship between the load at room temperature and the obtained pressure at low temperature. The data in this experiment coincides well with the data obtained by the previous pressure cell [4]. It is noted that the pressure generated in the sample

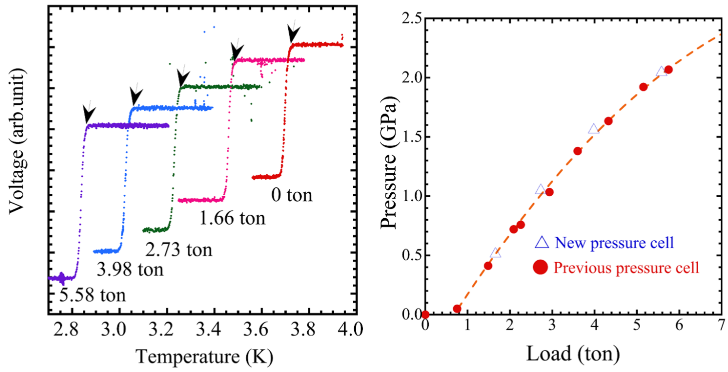

Fig. 2 (a) Temperature dependence of AC susceptibility of Sn at various loads. The arrows indicate the onset of the superconducting transition of Sn. (b) The relationship between the pressure at low temperature and the load at room temperature. The dotted line is the fitting line by the quadratic form.

space is unchanged basically unless the inner diameter of the cylinder is changed in spite of the outer diameter. Moreover, this result shows that the new cylinder can generate the pressure at least up to 2 GPa within the elastic deformation. As described above, we optimized the pressure cell which can generate the pressure at least up to 2 GPa repeatedly for the standard superconducting magnet with VTI.

The experimental setup for the ESR measurement is the same as that in the previous study. The details are found in Refs. [4,6]. Gunn oscillators and backward traveling wave oscillators (BWOs) are used. The frequency region is from 50 to 700 GHz. The insert pipe chamber is filled with a small amount of He exchange gas. The temperature region from 1.7 to 200 K is available.

3 Application to Quantum Magnet $CsCuCl_3$

As an application example of this new ESR system, we show the result on a triangular lattice antiferromagnet $CsCuCl_3$. $CsCuCl_3$ is an S =1/2 quantum magnet which forms the one-dimensional ferromagnetic chain along the *c*-axis and the two-dimensional antiferromagnetic triangular lattice in the *c*-plane [11]. It is well known that $CsCuCl_3$ shows field-induced phase transition below Néel temperature T_N = 10.7 K for $H \parallel c$ at $H_c \sim 12$ T [12]. The intensive investigation revealed that the spin configuration changes from the umbrella structure to the 2-1 coplaner structure at H_c and the quantum effect plays an important role to this phase transition [13]. We performed high-pressure ESR measurement of this compound and we observed a large pressure effect, though the field range was limited up to 10 T [14]. Recently, $CsCuCl_3$ was found to show an interesting phenomenon around the transition field H_c under high pressure [15]. Therefore, this compound is a good example to try our new ESR system which can reach this transition field.

A single crystal with a size of around 3 mm×3 mm×1 mm was used. We performed the ESR measurement at ambient pressure and at 1.42 GPa. The ex-

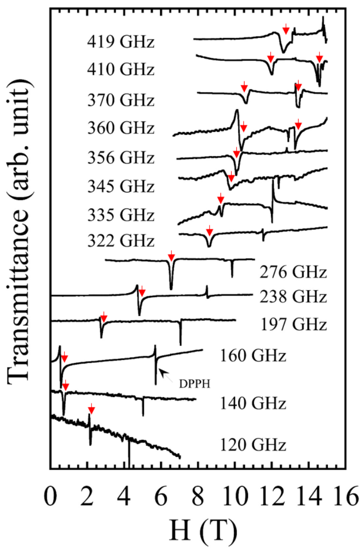

Fig. 3 Frequency dependence of ESR spectra of $CsCuCl_3$ for $H \parallel c$ at 1.7 K at 1.42 GPa.

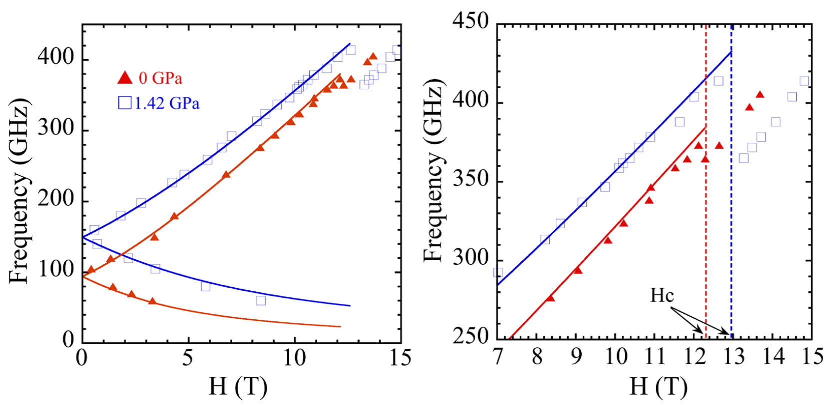

Fig. 4 Frequency field diagram of $CsCuCl_3$ for $H \parallel c$ at 1.7 K at 0 GPa and 1.42 GPa (a) and its enlarged view around H_c (b).

periment was done at 1.7 K in the frequency region from 60 GHz to 420 GHz. The field was applied parallel to the c-axis.

Figure 3 shows the frequency dependence of the ESR absorption spectra of $CsCuCl_3$ for $H \parallel c$ at 1.7 K. The pressure is estimated to be 1.42 GPa from the load at room temperature using the relationship shown in Fig. 2 (b). Only one absorption line is observed below 356 GHz, while another absorption line appears at the higher field side above 360 GHz. Figure 4 (a) is the frequency-field diagram at 1.42 GPa. We also plotted the data obtained at ambient pressure. The observed resonance mode is the antiferromagnetic resonance (AFMR) mode. It is clearly seen that the AFMR mode changes significantly as the pressure is increased. The solid lines below H_c are the AFMR mode for the umbrella structure obtained from the mean field approximation [16]. We used $g = 2.11$ as the g-value in the paramagnetic region, which hardly depends on the pressure up to 1.5 GPa at room temperature [5]. The fitting parameter is the antiferromagnetic gap at zero field and the gaps at 0 GPa and 1.42 GPa are obtained to be 94.2 GHz and 149.5 GHz, respectively. The antiferromagnetic gap energy is enhanced by the pressure and the tendency is quantitatively consistent with the previous results [14].

Figure 4 (b) is the enlarged view of the AFMR modes in the high field region. The discontinuous change of the AFMR mode is clearly seen. At ambient pressure, Ohta *et al.* found that this discontinuous change corresponds to the transition field H_c due to the change of the spin configuration [17]. The transition fields H_c are obtained to be 12.3 T and 13.0 T at 0 GPa and 1.42 GPa, respectively. The increase of the transition field was already observed by the magnetization measurement under pressure [15]. We confirmed the same tendency from the newly developed high-pressure ESR system. The magnetization measurement suggests that the spin configuration above H_c changes by applying the pressure. In order to observe such change from the microscopic point of view, this high-pressure ESR measurement is a promising tool and further investigation is required.

4 Summary

We have developed a high-field and high-pressure ESR system using a widely used Oxford 15 T superconducting magnet. We optimized new pressure cell to fit the VTI of the 15 T magnet. From the result of the pressure generating test, we confirmed that the pressure is attainable up to at least 2 GPa by new pressure cell. We applied this new ESR system to S = 1/2 triangular antiferromagnet $CsCuCl_3$. It was confirmed that the antiferromagnetic gap of $CsCuCl_3$ increases as the pressure is applied. It was also found that the transition field due to the change of the spin configuration increases as the pressure is applied.

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