神戸常盤大学紀要　創刊号　2009

報告

Production of polarized ^{3}He gas by means of very low temperature and high magnetic field

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極低温と強磁場による偏極^{3}He ガスの発生

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SUMMARY

Recently, we started a project on polarized ^{3}He gas generated by the brute force method with very low temperature (～4mK) and strong static magnetic field (B_{max}=17T), and rapid melting of highly polarized solid ^{3}He followed by gasification. The aim of this project is to use highly polarized ^{3}He as, so called, a hyperpolarized ^{3}He MRI (Magnetic Resonance Imaging) for medical application. A Pomeranchuk cell to be cooled by ^{3}He－^{4}He dilution refrigerators is designed and fabricated. For an initial test experiment, the Pomeranchuk cell will be installed to a low power ^{3}He－^{4}He dilution refrigerator (IBC; In Beam Cryostat) with a 1 T superconducting solenoidal coil.

On the other hand, it will be installed to a high power ^{3}He－^{4}He dilution refrigerator (DRS2500) with a 17T superconducting solenoidal coil for a full-scale experiment.

In this report, we present outline of the principle of polarizing ^{3}He , and a present status of design and construction of the experimental equipment.

要　旨

最近、我々は極低温（～ 4mK）と超強磁場（最大17T）を用いた所謂Brute force法による偏極固体^{3}Heの生成と急速解凍による偏極^{3}Heプロジェクトをスタートした。このプロジェクトの目的は、偏極^{3}Heガスをハイパー偏極^{3}He MRI（核磁気共鳴イメージング）に用いて医学に応用することである。^{3}He－^{4}He希釈冷凍器に取り付けられるポメンランチュークセルが設計され、製作されている。最初のテスト実験では、ポメランチュクセルは低い冷凍能力を有する^{3}He－^{4}He希釈冷凍器（IBC; In Beam Cryostat）に取り付けられる。そこでは、1Tの超伝導ソレノイドコイルが用いられる。

一方、本実験では、ポメランチュクセルは高い冷凍能力を有する^{3}He－^{4}He希釈冷凍器（DRS 2500）に取り付けられる。そこでは、17Tの超伝導ソレノイドコイルが用いられる。

このレポートで、我々は、^{3}Heを偏極させる原理と実験装置の設計・製作の現状を述べる。

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1. INTRODUCTION

The NMR (Nuclear Magnetic Resonance) is nowadays widely used for a probe in various scientific, technological, and medical researches. One of the advanced applications of the NMR is the MRI (Magnetic Resonance Imaging). In particular, the MRI has shown a remarkable usefulness in medical diagnostics as a complementary tool to the X-ray CT. Most of the MRI working clinically has been the proton MRI except for a few cases used for the laboratory base.

Recently, the MRI with highly polarized ^{3}He and ^{129}Xe gases, so called "Hyperpolarized MRI", became available on the laboratory base. Since this type of MRI observes signals from these gases, the lung imaging so far difficult to be observed due to the low density of tissues is considered to be possible.

On the basis of the above aspect, we have started developing a novel technique to efficiently generate a highly polarized ^{3}He gas for the Hyperpolarized ^{3}He MRI. Differing from the methods so far, in which an optical pumping was used for producing ^{3}He gas, our project is associated with a brute force method, in which a very low temperature and high magnetic field are employed. For this purpose we use a Pomeranchuk cooling cell connected to a dilution refrigerator and a superconducting magnet, thus realizing T～4mK and B = 17T.

In the following, we will touch an outline of the principle of polarization and the present state of design and construction of the experimental setup.

2. PRINCIPLE OF ^{3}HE POLARIZATION

2.1. Brute force method

Generally speaking, the Brute force method uses the polarization by a very low temperature and strong magnetic field. The nuclear polarization, P_N attainable with this method obeys the Curie's law and expressed in terms of temperature, T (K) and magnetic field, B (T) as

$$P_N = \tanh(x), \tag{1}$$

where x is given by

$$x = \frac{g_{3\mathrm{He}}\mu_N B}{2kT}, \tag{2}$$

where $g_{3\mathrm{He}}$ is a nuclear g-factor of ^{3}He, μ_N is a nuclear magneton, k is the Boltzman constant, respectively. In Fig. 1, the P_N is plotted as a function of x. It is simply evaluated that P_N exceeds 95% with T = 4mK, and B =17

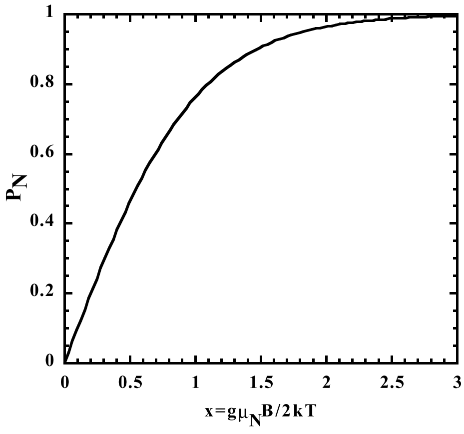

FIGURE 1. ^{3}He nuclear polarization plotted as a function of x

T. It is well known that ^{3}He is liquefied below 3.8K, but never solidified at normal pressure. As far as liquid ^{3}He is concerned, one cannot expect a large amount of polarization because liquid ^{3}He is a Fermi liquid, where only a minor part of the ^{3}He nuclei around the magnetic Fermi temperature （T^*_F = 180mK） can be polarized even when the brute-force condition is satisfied. As a result, the ^{3}He polarization for liquid ^{3}He could never exceed a % order.

About 30 years ago, Castaing and Nozières proposed[1] an idea to produce highly polarized ^{3}He liquid. Their idea was later proved experimentally[2,3]. The idea is to produce polarized solid ^{3}He in the first step, and, the solid ^{3}He is, then, liquefied. Here, it is noted that solid ^{3}He can be highly polarized since solid ^{3}He does not obey the Fermi statistics, and shows a paramagnetism under the presence of external magnetic field at $T >$ 1mK. If this principle can be extended to production of polarized ^{3}He gas, production of a large amount, say, ～1m^3/day of highly polarized （$\geq$ 80%） ^{3}He may be available. This attractive technique is what we are planning to produce a plenty amount of highly polarized ^{3}He gas.

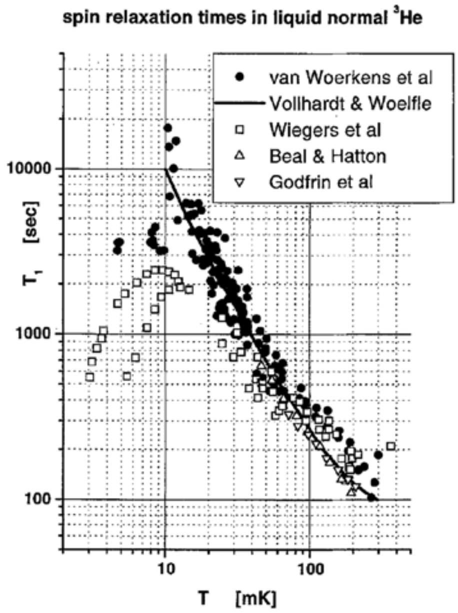

FIGURE 2. Observed relaxation times of ^{3}He in liquid phase

However, there exists a serious problem in ^{3}He liquid phase. It is well known that the spin relaxation time for ^{3}He in liquid phase is not long. Therefore, the liquid ^{3}He should be rapidly gasified within the relaxation time. In fact, a summary of the observed ^{3}He spin relaxation times are plotted as a function of temperature for liquid in Fig. 2[4]. Since the ^{3}He relaxation time becomes relatively short at around 300mK （$\leq$ 200s）, it is required to gasify liquid ^{3}He within the above time. This process is called "rapid melting", and an establishment of the rapid melting process is one of the most important goals for our project.

2.2. Pomeranchuk cooling

As mentioned in Subsec. , it is of particular importance to solidify liquid ^{3}He for attaining a high polarization. The solidification of liquid ^{3}He can be realized by compressing liquid ^{3}He . In Fig. 3, the melting curve of solid ^{3}He is shown, where ^{3}He pressure, P is plotted as a function of temperature, T. From this curve, it is necessary to compress liquid ^{3}He with a pressure typically larger than 3MPa at a temperature lower than 320mK.

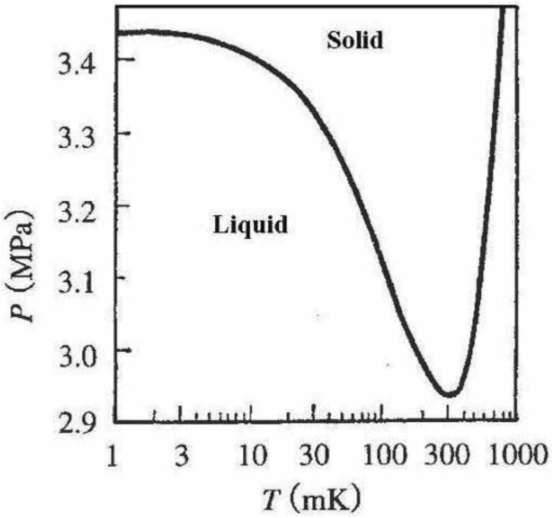

FIGURE 3. Melting curve of ^{3}He plotted as a function of temperature

Another importance of Fig. 3 is the fact that P increases according to decreasing T, i.e., the pressure gradient, $(\frac{dP}{dT})_{melt}$ has a negative value. This phenomenon is crucially important in understanding the principle of the Pomeranchuk cooling as mentioned below.

According to the equation of Clausius-Clapeyron, the pressure gradient is expressed in terms of entropies and volumes of liquid ^{3}He.

$$\left(\frac{dP}{dT}\right)_{melt} = \frac{S_L - S_S}{V_L - V_S}, \tag{3}$$

where S_L and S_S are an entropy of liquid ^{3}He and solid ^{3}He , respectively, and V_L and V_S are a volume of liquid ^{3}He and solid ^{3}He , respectively. Since $V_L > V_S$ in this temperature region, it is requested that $S_L < S_S$ from Eq. 3. In other words, the solid entropy becomes larger than the liquid one in this temperature region. In fact, entropies for liquid and solid ^{3}He phases are plotted as a function of temperature, T in Fig. 4. This result suggests us that the ^{3}He temperature is decreased by adiabatically compressing liquid ^{3}He . This is the principle of the Pomeranchuk cooling. Since the ^{3}He solidification is accompanied simultaneously by lowering ^{3}He temperature, it would be beneficial to increase the polarization according to eq. (1).

3. TEST EXPERIMENT WITH POMERANCHUK CELL

We have a plan to construct an equipment enabling rapid melting of solid ^{3}He by employing a ^{3}He－^{4}He dilution refrigerator (DRS2500: Leiden Cryogenics), and a 17-T superconducting magnet (17T71:Jastec)

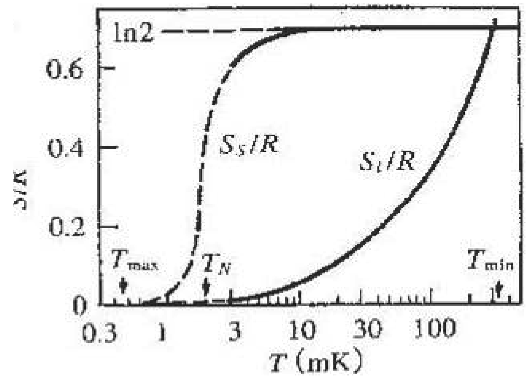

FIGURE 4. Entropies of liquid and solid ^{3}He

currently used for the polarized HD (Hydrogen Deuteride) target developed at RCNP.

It is of particular importance to make a test experiment before a full-size experiment in order to much learn about the low temperature physics because all of us are not well familiar with it. For this purpose, we decided to start with a ^{3}He − ^{4}He dilution cryogenic system which will be used for an experiment planned at SPring-8, Hyogo, Japan in near future. We name this cryostat IBC (In Beam Cryostat) which has refrigeration power down to 200mK. In Fig. 5, we show a drawing of IBC, where a 1 T superconducting solenoidal coil is attached. This means we can cool the Pomeranchuk cell down to a temperature lower than 250mK under the influence of an external magnetic field of 1 T. With this system, we can expect ^{3}He polarization of about 0.1% . Of course, this is far smaller than our final goal. However, this number is enough large to be detected as the test experiment.

When liquid ^{3}He temperature is lower than 320mK, it is, adiabatically, compressed by ^{3}He gas, and further compressed by a ^{4}He gas through a mechanical piston of the cooling cell which is called a Pomeranchuk cell. The designed and fabricated Pomeranchuk cell is shown in Fig. 6, where a) is the drawing and b) is the constructed Pomeranchuk, respectively. The creation of liquid and solid ^{3}He is done in a cell made of polycarbonate and fixed to a piston mechanics with Stycast 2850FT. A ruthenium oxide sensor is inserted in the polycarbonate cell for temperature measurement. A two turn NMR coil is attached outside the polycarbonate cell. To restrict thermal flow gas introducing tubes are made of cupronickel tube with an inner diameter of 0.2mm$^\phi$. An outer surface of the Pomeranchuk cell is gold-plated in order to reduce the heat flow due to radiation. To monitor the pressure inside the polycarbonate chamber used for compressing ^{3}He , a pressure sensor based on the capacitance measurement is used as shown in Fig. 7.

The main part of the sensor is made of Beryllium-copper alloy with a sensitive area consisting of a thin layer with 0.5mm in thickness. The space between two electrodes is chosen 20μ at room temperature. The measured capacitance at room temperature was 31 pF, which is consistent with the predicted value.

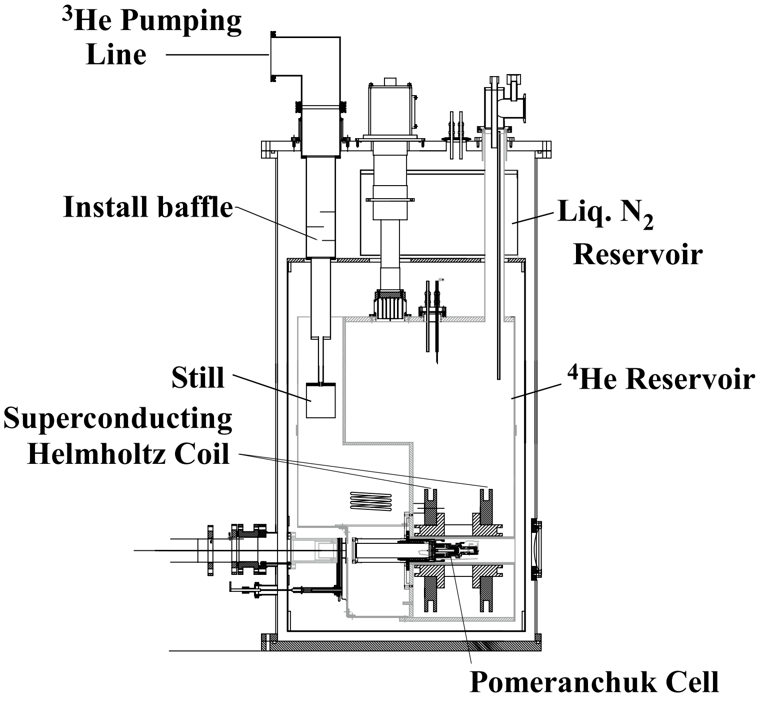

FIGURE 5. ^{3}He−^{4}He Dilution refrigerator used for checking the principle of Pomeranchuk cooling

Before starting with the DRS2500 cryostat, we are serving another dilution cryogenic system to be used as an inbeam cryostat (IBC), which has a cooling power down to 250mK for the purpose that we become familiar with the low temperature technology. This IBC has a 1-T superconducting Helmholtz coil as sketched in Fig. 5. Basic data needed for designing a practical device will be taken with this test system. ^{3}He gas in a cooling cell connected to a mixing chamber of the dilution refrigerator is cooled down and liquefied.

Gas pressures of ^{3}He and ^{4}He should be carefully controlled for successful Pomeranchuk cooling. For this purpose, we constructed a gas handling system as shown in Fig. 8, where the upper drawing is a system for controlling ^{3}He and the lower one is a system for controlling ^{4}He. Vacuum test and high pressure test ($\sim$ 3MPa) were performed, and found that the performance obtained by these test was acceptable for our first step experiment. The dip stick denoted in the upper picture of Fig. 8 works as a compressor of ^{3}He gas ($\geq$ 3.4MPa) by using charcoal powder cooled at liquid He temperature. In the future work, the Pomeranchuk cell will be thermally disconnected from the Leiden dilution refrigerator by a thermal switch5), and liquid ^{3}He is solidified and cooled down to a few mK by the principle of Pomeranchuk cooling6). When the magnetic field of 17-T is applied to the cooling cell, the ^{3}He nuclei are highly polarized according to eq. (1).

4. CONCLUSION

A novel method to produce a large amount of highly polarized ^{3}He gas was proposed. The method of the polarization is based on the brute force method, in which a low temperature ($T \leq 10$mK) and a high magnetic field ($B \sim 17$T). For this purpose, a dilution cryostat and a superconducting magnet, which are currently used for the project of the polarized HD target, are employed. Using a principle of the Pomeranchuk cooling, ^{3}He is solidified and cooled.

ACKNOWLEDGEMENT

The authors are grateful to Prof. T. Kishimoto, the RCNP director, and RCNP staff for their sincere encouragement. Many thanks are due to Prof. G. Frossati (Leiden, the Netherlands), Prof. J. P. Didelez (IPN, Paris, France), and H. Fujiwara (Medical School of Osaka University) for providing us many valuable information on the low temperature technology. This work has been financially supported mainly by the RCNP and HX-Pharma Corporation.

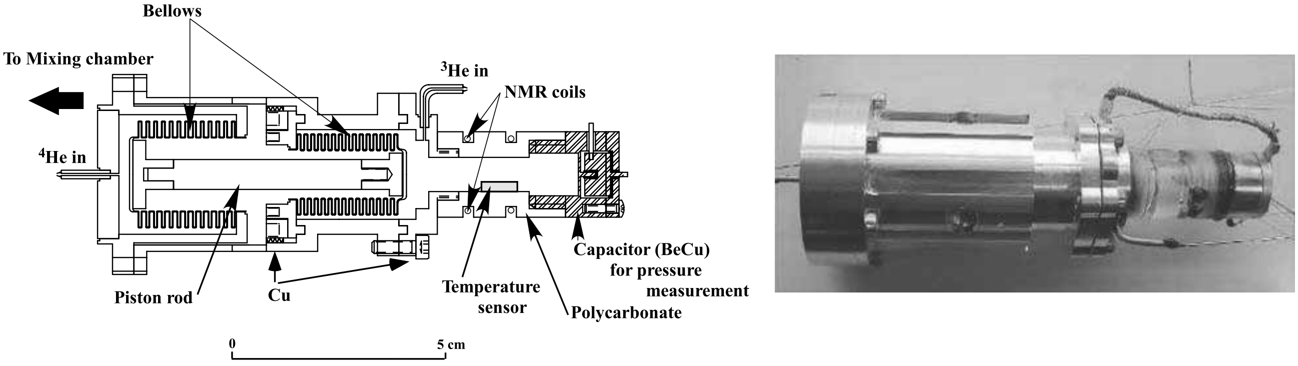

FIGURE 6. Drawing and photo of the Pomeranchuk cell constructed in the present work

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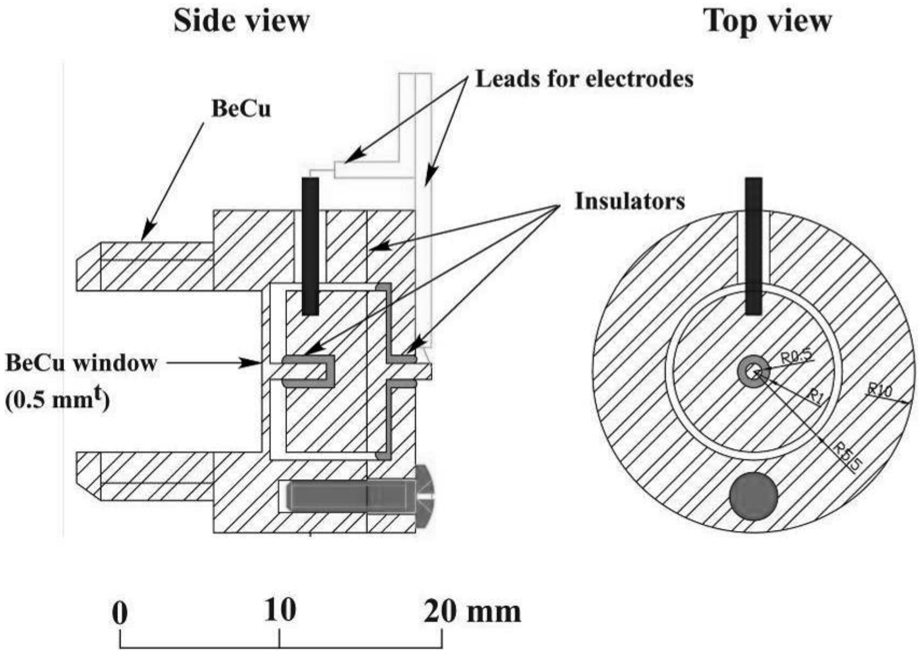

FIGURE 7. Pressure sensor based on a capacitance measuement

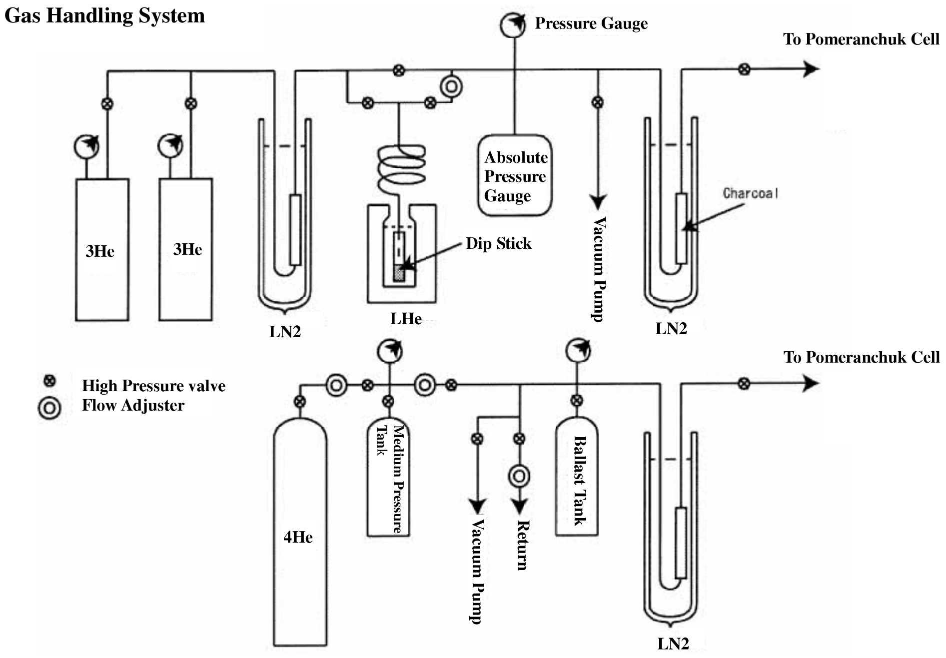

FIGURE 8. Gas handling system for Pomeranchuk cell. ^{3}He gas system (upper). ^{4}He gas system(lower).