

# Optically Polarized Alkali Metal Cell for Muonic Helium Measurements

Takashi INO<sup>1,2</sup>, Seiso FUKUMURA<sup>3</sup>, Patrick STRASSER<sup>1,2</sup>, Masaki FUJITA<sup>4</sup>, Yoichi IKEDA<sup>4</sup>, Sohtaro KANDA<sup>1,2</sup>, Masaaki KITAGUCHI<sup>3</sup>, Shoichiro NISHIMURA<sup>1,2</sup>, Takayuki OKU<sup>2,5</sup>, Takuya OKUDAIRA<sup>2,3,5</sup>, Hirohiko M. SHIMIZU<sup>3</sup>, and Koichiro SHIMOMURA<sup>1,2</sup>

<sup>1</sup>KEK, High Energy Accelerator Research Organization, Tsukuba 305-0801, Japan

<sup>2</sup>J-PARC Center, Tokai 319-1195, Japan

<sup>3</sup>Department of Physics, Nagoya University, Nagoya 464-8601, Japan

<sup>4</sup>Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

<sup>5</sup>JAEA, Japan Atomic Energy Agency, Tokai 319-1195, Japan

\*E-mail: [takashi.ino@kek.jp](mailto:takashi.ino@kek.jp)

(February 9, 2022)

An optical pumping cell was fabricated for studies of muonic helium hyperfine structure. The cell contained drops of Rb and K, 3.2 atm of <sup>4</sup>He, and 0.1 atm of N<sub>2</sub>. The alkali metal was optically polarized and determined by EPR to be nearly 100% polarization. Possible deterioration in the Pyrex glass cell was found after optical pumping at elevated temperatures up to ~200°C, probably due to the reaction with alkali metal. Another optical pumping cells made of alkali resistant aluminosilicate glass are necessary for further studies.

**KEYWORDS:** muonic helium, muon mass, muon magnetic moment, spin exchange optical pumping, alkali hybrid SEOP, alkali metal polarization

## 1. Introduction

Muonic helium is an atom in which one of the two electrons is replaced by a negative muon. The hyperfine structure of muonic helium atoms is related to the negative muon mass and magnetic dipole moment as its ground-state Hamiltonian in a static magnetic field  $\mathbf{B}$  can be given in the equation below [1].

$$\mathcal{H} = (-h\Delta\nu)\mathbf{I}_\mu \cdot \mathbf{J} + (\mu_B^e g_J)\mathbf{J} \cdot \mathbf{B} + (\mu_B^\mu g'_\mu)\mathbf{I}_\mu \cdot \mathbf{B} \quad (1)$$

Here,  $h$  is the Planck constant,  $\Delta\nu$  is the hyperfine structure interval,  $\mathbf{I}_\mu$  is the muon spin operator,  $\mathbf{J}$  is the electron total angular momentum operator,  $\mu_B^e$  is the electron Bohr magneton, and  $\mu_B^\mu$  is the muon Bohr magneton. Represented as  $g_J$  and  $g'_\mu$  are the  $g$  factors for the electron and muon in the muonic helium atom, respectively. Precise measurements of the muonic helium hyperfine structure provide not only the negative muon mass and magnetic dipole moment but also opportunities to test a three-body atomic system as well as the CPT invariance in comparison with the positive muon.

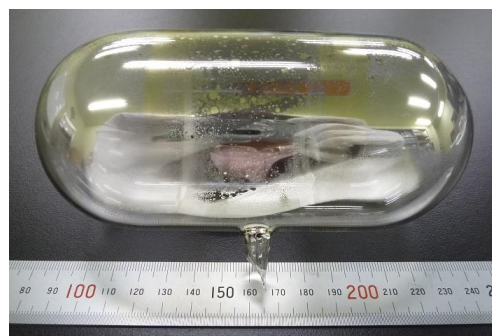
Muonic helium atoms can be formed simply by capturing muons in helium gas, but the polarization of initial muons, which is essential to measure the hyperfine structure, is largely lost in the process of muonic helium formation [2]. A clever technique to

*repolarize* muons in muonic helium atoms was demonstrated by Barton *et al.* [3]. They used optically-polarized alkali metal vapor in pressurized helium gas where the spins of unpaired electrons in alkali metal are exchanged with those of muons in muonic helium through the spin-exchange and charge-exchange collisions. This muon polarization method is similar to that for  $^3\text{He}$  nuclei known as spin-exchange optical pumping (SEOP), which is widely used in nuclear and particle physics, neutron scattering, magnetic resonance imaging, etc. [4].

We have proposed new precise measurements of muonic helium hyperfine structure at the J-PARC muon source [5, 6]. In the combination of the world's most intense pulsed muon beam [7] and the modern SEOP technology [8], the hyperfine structure of muonic helium will be determined with the lowest statistical uncertainty.

## 2. Hybrid Optical Pumping of Alkali Metal Atoms

In conventional  $^3\text{He}$  SEOP, Rb atoms are optically polarized, and the Rb polarization is transferred to that of  $^3\text{He}$  nuclei. It is known that most of the Rb polarization is lost by the Rb-Rb spin-destruction collisions [9, 10], and hence the optical power is predominantly wasted only to repolarize those Rb atoms. Babcock *et al.* [11] have developed an alkali-hybrid SEOP (AH-SEOP) method, which utilizes admixture of Rb and K instead of pure Rb in conventional SEOP. Still in AH-SEOP, only Rb atoms are optically polarized, but the Rb spins immediately spread to K atoms in the spin-exchange collisions between alkali metals. The beauty of AH-SEOP owes to much smaller K-K spin-destruction cross section while the spin-exchange cross section between K and  $^3\text{He}$  is roughly the same for Rb and  $^3\text{He}$  [10]. AH-SEOP can reduce the necessary optical power by a factor of 2 or more to keep the same alkali metal polarization [12]. This leads, compared to conventional pure Rb optical pumping (Rb-OP), twice as many alkali metal atoms can be polarized with the same optical power in alkali-hybrid optical pumping (AH-OP). Considering the similarity of  $^3\text{He}$  SEOP and the repolarization of muonic helium, it is likely that the spin-exchange and charge-exchange cross sections between K and muonic helium are comparable to those for Rb, and hence we expect the repolarization rate of muonic helium, which is proportional to the number density of polarized alkali metal atoms, can be increased by AH-OP. An essential condition in AH-OP to achieve the highest number density of polarized alkali metal atoms is the mixture ratio of K and Rb. Both K and Rb are in liquid state at nominal temperatures of optical pumping,  $\sim 200^\circ\text{C}$ , and the number density of alkali metal in gas phase is proportional to the vapor pressure that increases with temperature. The optimal condition for the K to Rb vapor density ratios in  $^3\text{He}$  AH-SEOP were studied to be  $2 - 6$  and  $5 \pm 2$  in Refs. [12, 13], respectively. The best K and Rb mixture in AH-OP for the muonic helium repolarization may be similar to that in AH-SEOP.

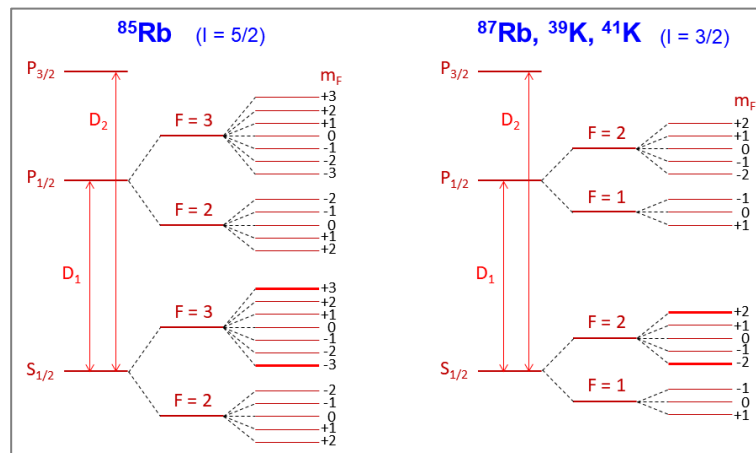


**Fig. 1.** Prototype  $^4\text{He}$  cell for muonic helium hyperfine structure measurements.

### 3. Alkali Metal Cell with Pressurized $^4\text{He}$ Gas

We prepared a prototype sealed glass cell filled with Rb, K,  $\text{N}_2$ , and  $^4\text{He}$  as pictured in Fig. 1. The glass cell was blown from a Pyrex borosilicate glass tube at the glass workshop of Institute of Multidisciplinary Research for Advanced Materials in Tohoku University. The cell was cylindrical with half-spheres at both ends. The dimensions were 72 mm in diameter and 154 mm in length. The volume was  $470\text{ cm}^3$ . The glass thicknesses were roughly 1.0 mm at the center of both ends and 1.8 mm in the cylindrical body.

The prototype  $^4\text{He}$  cell was fabricated in a similar manner to  $^3\text{He}$  AH-SEOP neutron spin filter (NSF) cells as described in Ref. [14]; first cleaned, then vacuum-baked for days, alkali metals chased into the cell, gas filled, and finally the cell was immersed in liquid nitrogen and sealed with a torch. Partial pressures of  $\text{N}_2$  and  $^4\text{He}$  gases were measured by a manometer at the time of sealing to be 0.1 and 3.2 atm at an equivalent temperature of  $0^\circ\text{C}$ , respectively. Note that a little  $\text{N}_2$  was added as the quenching gas for efficient optical pumping. The K to Rb mole ratio in the cell was determined  $\text{K/Rb} = 8.0$  by measuring the absorption strengths of white light at the  $\text{D}_1$  and  $\text{D}_2$  resonances of K and Rb for the  $^4\text{He}$  cell at temperatures between  $50$  to  $100^\circ\text{C}$  [12, 15]. The corresponding K to Rb vapor ratio became 1.3 at  $200^\circ\text{C}$  assuming each K or Rb vapor pressure obeyed Raoult's law, and the alkali metal number density was expected to be  $2 \times 10^{14}\text{ cm}^{-3}$ .



**Fig. 2.** Atomic energy levels for  $^{85}\text{Rb}$ , the nuclear spin  $I = 5/2$ , and the  $I = 3/2$  isotopes ( $^{87}\text{Rb}$ ,  $^{39}\text{K}$ , and  $^{41}\text{K}$ ). A total angular momentum and a magnetic sublevel of the atom are represented by  $F$  and  $m_F$ , respectively.

### 4. Optical Pumping Test

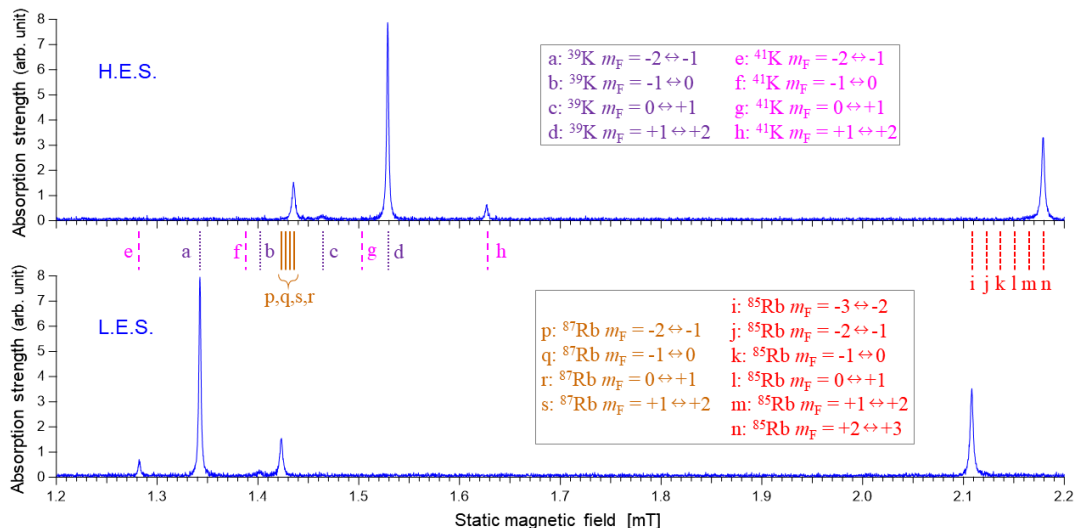
Figure 2 shows the atomic energy level diagrams of the Rb and K stable isotopes. Under optical pumping, the  $m_F = +3$  or  $-3$  sublevel in the  $S_{1/2}$  ground state is populated for  $^{85}\text{Rb}$  and  $m_F = +2$  or  $-2$  for the other isotopes (shown in bold lines in Fig. 2), depending on the circularity of the pumping light, but some atoms occupy other sublevels if alkali metal is not fully polarized. The polarization of alkali metal is determined from the occupancy in each sublevel. Electric paramagnetic resonance spectroscopy (EPR) can be used to determine the alkali metal polarization by measuring the relative populations in the  $m_F$  sublevels. The EPR signal strength is proportional to

the populations in adjacent two Zeeman sublevels whose transition energy corresponds to the EPR frequency in a static magnetic field.

An optical pumping test was performed for the prototype  $^4\text{He}$  cell to evaluate alkali metal polarization. The  $^4\text{He}$  cell was placed in a cylindrical aluminum oven, which was wrapped with a thin non-magnetic polyimide heater [16], and heated at  $200^\circ\text{C}$ . A non-magnetic thermocouple (copper-constantan) was attached to the  $^4\text{He}$  cell surface to monitor the temperature. The oven was placed in a static magnetic field inside a solenoid. A frequency-narrowed diode laser with an output power of 75 W was used for optical pumping of the  $^4\text{He}$  cell with a circularly polarized light. The oven, solenoid, and diode laser were similar to those used for a  $^3\text{He}$  NSF described in Ref. [8]. A small coil was set near the  $^4\text{He}$  cell at the opposite side of the diode laser to provide a 10 MHz radio-frequency field for EPR measurements. A photodiode was used to measure transmission intensity of the pumping laser through the  $^4\text{He}$  cell. The static magnetic field was swept through the transition resonances by ramping up/down the solenoid current. During the static magnetic field sweep, the absorption of the pumping laser increased when the EPR condition was met at any transition energy of the isotopes, and it led to a change of the photodiode signal. Alkali metal atoms were polarized either in the high-energy state (H.E.S.), which corresponded to  $m_F > 0$ , or the low-energy state (L.E.S.),  $m_F < 0$ , depending on the circularity of the pumping laser.

Figure 3 shows the absorption strength of the pumping laser in the  $^4\text{He}$  cell together with the calculated transition resonances for the Rb and K isotopes in vertical lines. Absorption peaks are clearly seen at the farthest transition resonances,  $m_F = \pm 3 \leftrightarrow \pm 2$  for  $^{85}\text{Rb}$  and  $m_F = \pm 2 \leftrightarrow \pm 1$  for the rest spin 3/2 isotopes. Each peak height relates to the natural abundance of an isotope [17]. There are small peaks noticeable at the transitions  $m_F = 0 \leftrightarrow +1$  for  $^{39}\text{K}$  in H.E.S and  $m_F = -1 \leftrightarrow 0$  in L.E.S. This fact indicates a small portion of  $^{39}\text{K}$  was in the  $m_F = +1$  or  $-1$  substate. The other isotopes also must have the second peaks but too small to be distinguished from the noise. The alkali metal in the  $^4\text{He}$  cell was not fully polarized but close to 100%.

For further studies, we intended to increase the cell temperature to boost the alkali metal number density but noticed that the prototype  $^4\text{He}$  cell had turned brown, probably due to the reaction with alkali metal. Such change of color has never been observed at these temperatures for  $^3\text{He}$  NSF cells, which are made of aluminosilicate glass. Pyrex glass can be used for Rb-OP at lower temperatures but not for AH-OP.



**Fig. 3.** Absorption strengths of the pumping laser in the  $^4\text{He}$  cell with alkali metal polarized in the high-energy state (top) and the low-energy state (bottom). The transition resonances for the K and Rb isotopes are shown in vertical lines.

## 5. Conclusions

We made a prototype  $^4\text{He}$  cell for studies of muonic helium hyperfine structure. The alkali metal in the  $^4\text{He}$  cell was optically polarized at  $200^\circ\text{C}$  and determined by EPR to have nearly 100% polarization. The alkali metal number density in the  $^4\text{He}$  cell was expected to be  $2 \times 10^{14} \text{ cm}^{-3}$  from the vapor pressure of alkali metal at the optical pumping temperature. Further studies are necessary to optimize the mixture of Rb and K to maximize the number density of polarized alkali metal atoms.

Aluminosilicate glass named GE-180 is the de facto standard material used for  $^3\text{He}$  NSF cells everywhere. To our knowledge, it does not change color under optical pumping up to  $\sim 250^\circ\text{C}$ . The only drawback of this glass is glassblowing. GE-180 is much harder to work with than Pyrex borosilicate glass. This is the reason we fabricated the prototype  $^4\text{He}$  cell with Pyrex glass. However now, we have no choice but to use GE-180 for the muonic helium measurements. Another important aspect about the  $^4\text{He}$  cell is the window thickness. Muons captured in the window material will produce background in the measurements. The background should be minimized while the safety of the pressurized  $^4\text{He}$  cell must be ensured.

## Acknowledgment

We would like to thank Mr. Tomomi Kudo of Tohoku University for the glass cell blowing. This work was performed partially under the GIMRT Program of the Institute for Materials Research, Tohoku University (Proposal No. 20N0007). This work was also supported financially in part by JSPS KAKENHI Grants Nos. 16H02125 and 20H05646.

## References

- [1] P. A. Souder *et al.*, Phys. Rev. A **22**, 33 (1980).
- [2] V. W. Hughes and G. zu Putlitz, in *Quantum Electro-dynamics*, ed. T. Kinoshita (World Scientific, Singapore, 1990) p. 822.
- [3] A. S. Barton *et al.*, Phys. Rev. Lett. **70**, 758 (1993).
- [4] T. R. Gentile, P. J. Nacher, B. Saam, and T. G. Walker, Rev. Mod. Phys. **89**, 045004 (2017).
- [5] P. Strasser, K. Shimomura, and H. A. Torii, JPS Conf. Proc. **21**, 011045 (2018).
- [6] S. Fukumura *et al.*, “Proposal for new measurements of muonic helium hyperfine structure using SEOP at J-PARC,” Proceedings of International Conference on Exotic Atoms and Related Topics (EXA 2021), submitted.
- [7] N. Kawamura *et al.*, J. Phys.: Conf. Ser. **551**, 012062 (2014).
- [8] T. Ino *et al.*, J. Phys.: Conf. Ser. **862**, 012011 (2017).
- [9] A. Ben-Amar Baranga *et al.*, Phys. Rev. Lett. **80**, 2801 (1998).
- [10] R. J. Knize, Phys. Rev. A **40**, 6219 (1989).
- [11] E. Babcock *et al.*, Phys. Rev. Lett. **91**, 123003 (2003).
- [12] W. C. Chen, T. R. Gentile, T. G. Walker, and E. Babcock, Phys. Rev. A **75**, 013416 (2007).
- [13] J. T. Singh *et al.*, Phys. Rev. C **91**, 055205 (2015).
- [14] T. Okudaira *et al.*, Nucl. Instrum. Methods Phys. Res. A **977**, 164301 (2020).
- [15] The resonance wavelengths for K and Rb in vacuum. K-D<sub>1</sub> : 770.108 nm, K-D<sub>2</sub> : 766.701 nm, Rb-D<sub>1</sub> : 794.979 nm, Rb-D<sub>2</sub> : 780.241 nm.
- [16] T. Ino, H. Hayashida, H. Kira, T. Oku, and K. Sakai, Rev. Sci. Instrum. **87**, 115198 (2016).
- [17] Natural abundances of K and Rb isotopes.  $^{39}\text{K}$  : 93.3%,  $^{41}\text{K}$  : 6.7%,  $^{85}\text{Rb}$  : 72.2%,  $^{87}\text{Rb}$  : 27.8%.