CHIN. PHYS. LETT. Vol. 38, No. 9 (2021) 092801

Development of a Spin-Exchange Optical Pumping-Based Polarized ^{3}He System at the China Spallation Neutron Source (CSNS)

Chuyi Huang(黄楚怡)[1,2†], Junpei Zhang(张俊佩)[1,2†], Fan Ye(叶凡)[1,2], Zecong Qin(秦泽聪)[1,2], Syed Mohd Amir[1,2], Zachary Norris Buck[1,2], Ahmed Salman[1,2], Wolfgang Kreuzpaintner[1,2], Xin Qi(齐欣)[1,2], Tianhao Wang(王天昊)[1,2*], and Xin Tong(童欣)[1,2*]

[1]*Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China*
[2]*Spallation Neutron Source Science Center, Dongguan 523803, China*

(Received 7 June 2021; accepted 13 July 2021; published online 2 September 2021)

Polarized ^{3}He neutron spin filters (NSFs) can be used as a vital tool for neutron polarization production and analysis. The China Spallation Neutron Source (CSNS), as one of the major neutron facilities in China, has committed resources to the development of a polarized ^{3}He NSF program to support its growing polarized neutron research. A spin-exchange optical pumping (SEOP)-based polarized ^{3}He system and other necessary hardware for NSF transport has been recently developed. The performance of the system is benchmarked using an in-house developed cell named "Trident". Neutron beam measurements yield a ^{3}He polarization of 77% with over 200 h of on-beam relaxation time. Combining this newly developed SEOP system with the recently reported cell fabrication station, CSNS is now capable of the fully self-sustained production of ^{3}He NSFs that shall support its future neutron polarization research.

DOI: 10.1088/0256-307X/38/9/092801

Neutron scattering is an invaluable technique capable of investigating both the structure and dynamics of a wide variety of materials.[1–3] Spin-polarized neutrons can further provide unique information about material properties, which may be inaccessible using other scattering techniques. Typical applications[4–7] provide powerful tools for the study of complex magnetic materials from atomic to macroscopic scale. Consequently, polarized neutron techniques constitute an essential building block of world-leading neutron research facilities.

The ^{3}He gas container,[8,9] also known as a neutron spin filter (NSF) cell, utilizes the strongly spin-dependent absorption cross-section of spin-polarized ^{3}He gas so that the neutron beam will be partiality absorbed (i.e. filtered) as it traverses the device. Compared with the polarizing monochromators[10] and supermirrors,[11] the ^{3}He NSF allows for a fast and quasi-zero alignment in the neutron beam that makes NSFs highly attractive as polarizing devices, particularly, at spallation-based neutron sources. Because NSFs can also cover a large angular divergence,[12] NSFs are unrivaled in their applications as spin analyzing devices.

The achievable neutron polarization $P_{\rm n}$ of neutron transmission through the ^{3}He NSF is given by[8]

$$P_{\rm n} = \tanh(n\sigma l P_{^3{\rm He}}), \tag{1}$$

where $n\sigma l$ denotes the opacity of the NSF. Herein, n is the number density of the ^{3}He, $\sigma = \sigma_0\lambda$ with σ_0 being the ^{3}He absorption cross section for $\lambda = 1$ Å, λ is the neutron wavelength, l is the length of the neutron path through the NSF, and $P_{^3{\rm He}}$ is the ^{3}He polarization.

The application of polarized ^{3}He has become an integral part of research at neutron sources.[13–18] The CSNS,[19] as a newly commissioned neutron source, is developing and establishing its own SEOP ^{3}He program to supply polarized neutron capability to existing and future neutron instruments. In this manuscript, we report on the design and application of the recently developed ^{3}He SEOP station and the *off situ* NSF at the CSNS.

Design of the SEOP Station. SEOP is achieved by first polarizing unpaired electrons of alkali metal vapor coexisting with the ^{3}He gas. The spin-angular momentum is then transferred from the unpaired electrons to the ^{3}He nuclei via hyperfine interaction, thereby producing polarized ^{3}He gas. The SEOP station at the CSNS adopted the state-of-the-art method of mixing rubidium and potassium to reach a very high

Supported by the National Key Research and Development Program of China (Grant No. 2020YFA0406000), the National Natural Science Foundation of China (Grant No. 11875265), the Scientific Instrument Developing Project of the Chinese Academy of Sciences (Grant No. 284(2018)), Guangdong Basic and Applied Basic Research Foundation (Grant No. 2019B1515120079) and Dongguan Introduction Program of Leading Innovative and Entrepreneurial Talents (Grant No. 20191122).
†These authors contributed equally to this work.
*Corresponding authors. Email: wangtianhao@ihep.ac.cn; tongxin@ihep.ac.cn
© 2021 Chinese Physical Society and IOP Publishing Ltd

pumping efficiency.[20]

To implement such a process, the SEOP station at CSNS has been designed with five major components: a heating system, a high-power laser system, a magnetic field environment, a nuclear magnetic resonance (NMR) system and a transportation solenoid. A picture of the SEOP *off situ* station setup is shown in Fig. 1.

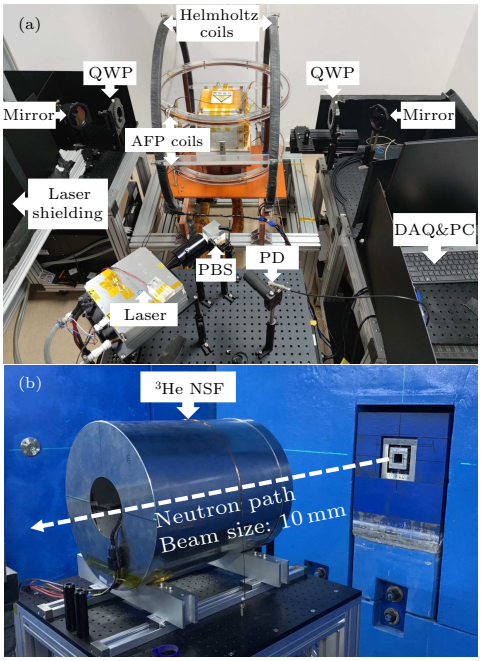

Fig. 1. A picture of the SEOP *off situ* station setup including (a) the laser lab and (b) the ^{3}He NSF.

Heating System. The spin exchange process occurs when the alkali metals are in their vapor phase with a suitable number density, which typically requires a temperature up to 200 ℃. To satisfy this requirement, the SEOP station at CSNS forces hot air from a heated compressed gas line. After 5 h of ramp-up time, the oven stabilizes the NSF temperature at a preset of 200 ℃ with fluctuations of approximately ± 1 ℃.

Laser System. The laser system on the SEOP station provides a high-power circularly polarized laser beam that polarizes the alkali metal.

For the developed SEOP station, a double-sided pumping laser optic is designed to provide a homogeneously illuminated area up to a diameter of ∼10 cm. The general layout is shown in Fig. 2, where a fiber coupled 160 W diode laser[21] with a central output wavelength of $\lambda = 794.8$ nm and a bandwidth of $\Delta\lambda \leq 0.3$ nm is used as the pumping source. The divergent unpolarized laser is parallelized by an adjustable convex lens. Following the lens, a polarization beam splitter (PBS) is applied to split the laser beam into two orthogonal linearly polarized branches. These two beams are guided from opposite sides through the center of the NSF by a pair of high-reflectivity coated silicon wafers, respectively. Before reaching the NSF, the polarization of the laser is converted into counter-circular polarization by a quarter wave plate (QWP) in each of the branches. The bi-directional pumping design, resulting in a coincide beam path and forming the shape of an isosceles right triangle (Fig. 2), contributes additional circular polarization momentum for pumping the alkali metal atoms.

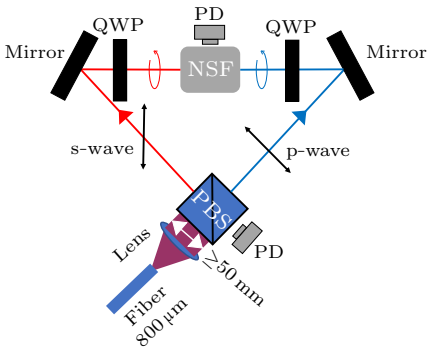

Fig. 2. Schematic of the laser system realized for the optical SEOP station.

Magnetic Field System. For reaching and maintaining a highest possible ^{3}He polarization, the external magnetic field must be homogeneous with a gradient not exceeding an order of magnitude of 10^{-4} cm^{-1} at the cell position.[22,23]

To meet these requirements, a pair of Helmholtz coils is used to generate the external magnetic field. The common axis of the coils aligns with the laser beam path. The coils generate a uniform magnetic field of $\sim 14 \times 10^{-4}$ T.

To verify the uniformity of the magnetic field in the cell region, magnetic field mapping was performed by a triple-axis Gaussmeter installed on a three-dimension step motor system.

Figures 3(a) and 3(b) demonstrate the absolute magnetic field and the field gradients along the x-axis. Typical magnetic field gradients of 5.8×10^{-4} cm^{-1} are obtained across the central region in which the NSF is located. The field uniformity is comparable with the field uniformity in similar systems reported elsewhere.[24]

Characterization and Spin Manipulation Systems. For monitoring the status of the SEOP process and for measuring the ^{3}He polarization, several NMR-based characterization and spin-flipping methods are implemented, which include the free induction decay (FID), adiabatic fast passage (AFP) and electron paramagnetic resonance (EPR).

The FID measurement provides a dynamic monitoring of the ^{3}He polarization through measuring the Larmor precession of the ^{3}He polarization within the applied magnetic field.[25] The AFP process flips the ^{3}He polarization with respect to its main holding mag-

netic field.[26,27] The EPR operation measures the absolute polarization of the ^{3}He inside the NSF cell through the interactions between the magnetic moment of ^{3}He nucleus and the spin of the alkali metal valence electron.[28] In combination with the AFP spin-flip capability, the resonance frequencies for the non-spin-flipped and spin-flipped states of the NSF can be obtained.

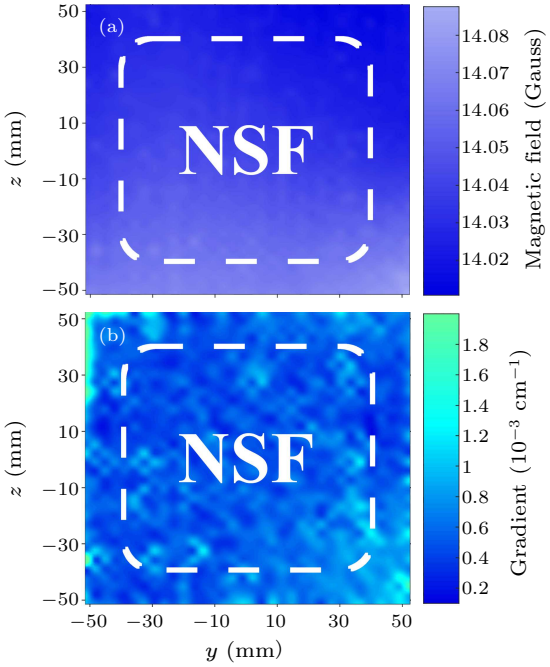

Fig. 3. Magnetic field mapping results for the central region in which the NSF is located. The absolute magnetic fields along the horizontal (x-axis) direction are given in (a). The total magnetic field gradient slices along the horizontal (x-axis) are shown in (b). The measured magnetic field gradient is $\sim 5.8 \times 10^{-4}$ cm^{-1}.

Transportation Solenoid. The SEOP station at CSNS is located at the laser lab within the CSNS target station, which allows an easy transfer of the polarized NSF onto the neutron beamline. The polarized NSF cells are preserved within a cylindrical transportation solenoid (36.5 cm in diameter, 42.0 cm in length). The transportation coil consists of two concentric coils sets and a μ-metal shield to provide a guide field with a uniformity up to 4×10^{-4} cm^{-1}. The transportation solenoid is also equipped with an FID coil attached to the contained cell surface to allow constant monitoring of the ^{3}He polarization. A set of sinusoidal coils are also installed to serve as the AFP coil mechanism to allow ^{3}He polarization to be flipped during the application of the NSF cells.

Results. The developed SEOP ^{3}He station was calibrated and characterized at the neutron technology development beamline (BL-20) at the CSNS. The CSNS BL-20 is a time-of-flight beamline that provides a wavelength band from 1 Å to 5.5 Å. Two boron carbide slits were installed to generate a 10-mm-diameter neutron beam that traversed the center of the NSF cell. For the calibration experiment, an in-house produced cell "Trident" was used as the NSF.[29]

The calibration experiment was performed by first determining the pressure of Trident by measuring the neutron transmission through unpolarized ^{3}He gas. The cell was then fully polarizing using the *off situ* SEOP station. During the pumping process, FID measurements were performed to monitor changes in the ^{3}He polarization, whereas EPR and AFP measurements were conducted to observe the real time ^{3}He polarization. The saturated cell was then transported to the BL-20 for neutron transmission measurements to quantify the polarization of the ^{3}He gas. Consecutive neutron transmission measurements were performed throughout a period of 11 days to monitor the lifetime of the Trident cell ^{3}He polarization.

Pressure of Trident. The pressure of the Trident was determined by neutron transmission measurements according to the following equation:[30]

$$T_0 = T_e \exp(-n\sigma l) = T_e \exp(-0.072 p\lambda \times l), \quad (2)$$

where T_0 is the neutron transmission for the unpolarized ^{3}He, T_e is transmission through empty glass, p is ^{3}He pressure in bar, and l is the path length inside the cell in cm. By plotting the neutron transmission as a function of wavelength and fitting to Eq. (2), the measurement yields a ^{3}He pressure of 1.58 bar at room temperature (25 ℃).

Polarization of ^{3}He Gas in Trident. The Trident was polarized on the developed SEOP station and reached saturation polarization after 15 h. Upon saturation of the ^{3}He polarization pumping, the ^{3}He polarization was flipped using the AFP method with an average loss of 0.49% through 10 consecutive flips.

The absolute ^{3}He polarization at the saturation was then characterized through an EPR measurement using the commonly recognized method.[31] The EPR measurement yielded a frequency shift of $2\Delta\nu = 15.8 \pm 0.5$ kHz under 200 ℃ in the SEOP station, which corresponds to a ^{3}He polarization of $P_{^3\mathrm{He}} = 77.7 \pm 2.5\%$.

The saturated ^{3}He polarization was then verified at BL-20. Comparing the neutron transmission of the polarized (T_n) and unpolarized cell (T_0), the ^{3}He polarization can be expressed as

$$T_n = T_0 \cosh(n\sigma l P_{^3\mathrm{He}}). \quad (3)$$

The measured neutron transmission is plotted as a function of neutron wavelength and fitted to Eq. (3), as shown in Fig. 4(b), which yielded a $P_{^3\mathrm{He}}$ of 77.4 ± 0.2%. The result shows that the polarization of ^{3}He calculated from the neutron transmission is consistent

with that of the EPR measurement on the SEOP station, while a negligible ^{3}He polarization is lost during the transport process.

Lifetime of Trident. Once the pumping for an NSF is terminated, the initial polarization $P^0_{^3\mathrm{He}}(t=0)$ decays with time t:

$$P_{^3\mathrm{He}}(t) = P^0_{^3\mathrm{He}} \exp\Big(-\frac{t}{T_1}\Big). \tag{4}$$

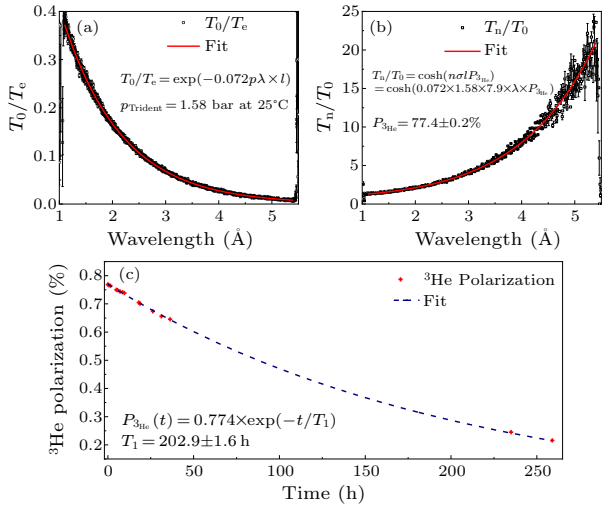

Fig. 4. Results of neutron transmission measurement. (a) Neutron transmission ratio of the unpolarized Trident to an empty cell resulting in a ^{3}He pressure of 1.58 bar at room temperature (25°C). (b) Neutron data of the fully polarized Trident. The polarization fitted by neutron transmission is $77.4 \pm 0.2\%$. (c) The decay curve of ^{3}He polarization of Trident. Its ^{3}He polarization was fitted from neutron transmission.

The lifetime T_1 of Trident ^{3}He polarization was measured through neutron transmission at the beginning and end of the 11-day (264 h) experiment. Once the polarization of the ^{3}He was measured for the first 40 h, we proceeded to other polarized neutron measurements while preserving the ^{3}He polarization within its transportation solenoid. The remaining ^{3}He polarization was once again measured at the end of the experiment with no other instruments on BL-20. Each measurement was analyzed identically through Eq. (3) to determine the ^{3}He polarization at the time of data collection. The decay of the ^{3}He polarization was then characterized by plotting the measured ^{3}He polarization as a function of time, as shown in Fig. 4(c). The fitted exponential decay revealed a relaxation time over 200 h (201.3 ± 11.8 h in SEOP station), which demonstrates the ability of the *off situ* system and transportation solenoid to maintain a high ^{3}He polarization of the NSF for use on CSNS beamlines.

Neutron Beam Polarization. For spallation neutron source, the neutron polarization $P_\mathrm{n} = \sqrt{1 - T_0^2/T_\mathrm{n}^2}$ can be deduced from the polarized and unpolarized NSF transmission according to Eqs. (1) and (3). A figure of merit (FOM) Q is given by $Q = T_\mathrm{n} \cdot {P_\mathrm{n}}^2$ to define the weight between neutron polarization and transmission for an NSF.[30] The results in Fig. 5 show that over 90% of neutron polarization above 2.12 Å can be produced by the 77% ^{3}He polarization, which also results in a neutron transmission of over 30% at 2 Å.

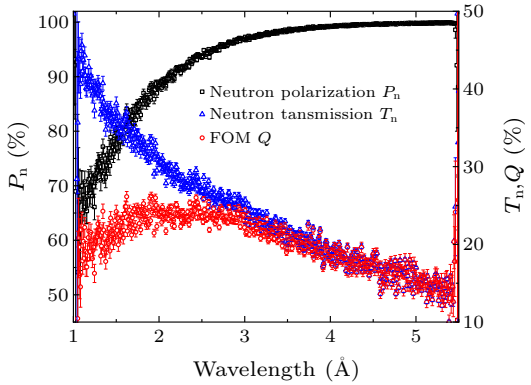

Fig. 5. Performance of Trident: Neutron polarization P_n, transmission T_n, and figure of merit Q as a function of neutron wavelength.

In conclusion, the results show that the SEOP system can efficiently pump the ^{3}He polarization of the "Trident" up to 77% within 15 h, which can operate with per AFP loss of ~0.5%. The transport system can be used to successfully deliver an *off situ* NSF to the neutron beamline (polarization loss during transport <0.3%) and maintain over 200 h of ^{3}He polarization relaxation time. This *off situ* NSF exhibits a preferable Q factor around 2 Å, from which over 90% neutron polarization at 2.12 Å and over 30% neutron transmission at 2 Å can be produced.

The SEOP station and *off situ* NSF marks the start of the neutron polarization era at CSNS. It will allow the polarized neutron capabilities to be delivered to neutron beamlines and promote further development of polarized ^{3}He NSFs in China.

We give special thanks to our glassblower Junsong Xie, who has been supportive in this project since the beginning. We are grateful to Dr. Songlin Wang for his guidance during the neutron tests on BL-20 at the CSNS. We also thank Michael Souza from Princeton University, United States of America, for his generous advice and technical help on our work.

References

[1] Wollan E O, Davidson W L and Shull C G 1949 *Phys. Rev.* **75** 1348
[2] Shull C G, Strauser W A and Wollan E O 1951 *Phys. Rev.* **83** 333
[3] Yarnell J L, Warren J L, Wenzel R G and Koenig S H 1964 *IBM J. Res. Dev.* **8** 234

[4] Hearmon A J, Fabrizi F, Chapon L C, Johnson R D, Prabhakaran D, Streltsov S V, Brown P J and Radaelli P G 2012 *Phys. Rev. Lett.* **108** 237201
[5] Cook J C, Richter D, Scharpf O, Benham M J, Ross D K, Hempelmann R, Anderson I S and Sinha S K 1990 *J. Phys. Condens. Matter* **2** 79
[6] Treimer W 2014 *J. Magn. Magn. Mater.* **350** 188
[7] Wang T, Jiang C Y, Bilheux H Z, Dhiman I, Bilheux J C, Crow L, McDonald L, Robertson L, Kardjilov N, Pynn R and Tong X 2019 *Rev. Sci. Instrum.* **90** 033705
[8] Coulter K P, Chupp T E, McDonald A B, Bowman C D, Bowman J D, Szymanski J J, Yuan V, Cates G D, Benton D R and Earle E D 1990 *Nucl. Instrum. Methods A* **288** 463
[9] Tong X, Jiang C Y, Lauter V, Ambaye H, Brown D, Crow L, Gentile T R, Goyette R, Lee W T, Parizzi A and Robertson J L 2012 *Rev. Sci. Instrum.* **83** 075101
[10] Freund A, Pynn R, Stirling W G and Zeyen C M E 1983 *Physica B+C* **120** 86
[11] Mezei F 1976 *Commun. Phys. (London)* **1** 81
[12] Chen W C, Gentile T R, Fu C B, Watson S, Jones G L, McIver J W and Rich D R 2011 *J. Phys.: Conf. Ser.* **294** 012003
[13] Boag S, Babcock E, Andersen K H, Becker M, Charlton T R, Chen W C, Dalgliesh R M, Elmore S D, Frost C D, Gentile T R, Lopez A R, Parnell S R, Petoukhov A K, Skoda M W A and Soldner T 2009 *Physica B* **404** 2659
[14] Gentile T R, Chen W C, Jones G L, Babcock E and Walker T G 2005 *J. Res. Natl. Inst. Stand. Technol.* **110** 299
[15] Jiang C Y, Tong X, Brown D R, Glavic A, Ambaye H, Goyette R, Hoffmann M, Parizzi A A, Robertson L and Lauter V 2017 *Rev. Sci. Instrum.* **88** 025111
[16] Okudaira T, Oku T, Ino T, Hayashida H, Kira H, Sakai K, Hiroi K, Takahashi S, Aizawa K, Endo H, Endo S, Hino M, Hirota K, Honda T, Ikeda K, Kakurai K, Kambara W, Kitaguchi M, Oda T, Ohshita H, Otomo T, Shimizu H M, Shinohara T, Suzuki J and Yamamoto T 2020 *Nucl. Instrum. Methods A* **977** 164301
[17] Salhi Z, Babcock E, Bingöl K, Bussmann K, Kammerling H, Ossovyi V, Heynen A, Deng H, Hutanu V, Masalovich S, Voigt J and Ioffe A 2019 *J. Phys.: Conf. Ser.* **1316** 012009
[18] Yan S, Zhang M F, Guo W C, Wang W Z, Gong J, Liang T J, Liu B Q, Peng M, Peng S M, Sun G, Tu X Q, Yan H Y, Zhang J H and Zheng H 2019 *Sci. Chin. Phys. Mech. Astron.* **62** 102021
[19] Chen H and Wang X L 2016 *Nat. Mater.* **15** 689
[20] Babcock E, Nelson I, Kadlecek S, Driehuys B, Anderson L W, Hersman F W and Walker T G 2003 *Phys. Rev. Lett.* **91** 123003
[21] https://www.qpclasers.com/
[22] McIver J W, Erwin R, Chen W C and Gentile T R 2009 *Rev. Sci. Instrum.* **80** 063905
[23] Cates G D, Schaefer S R and Happer W 1988 *Phys. Rev. A* **37** 2877
[24] Jiang C Y, Tong X, Brown D R, Culbertson H, Graves-Brook M K, Hagen M E, Kadron B, Lee W T, Robertson J L and Winn B 2013 *Rev. Sci. Instrum.* **84** 065108
[25] Parnell S R, Woolley E B, Boag S and Frost C D 2008 *Meas. Sci. Technol.* **19** 045601
[26] Babcock E, Petoukhov A, Chastagnier J, Jullien D, Lelièvre-Berna E, Andersen K H, Georgii R, Masalovich S, Boag S, Frost C D and Parnell S R 2007 *Physica B* **397** 172
[27] McKetterick T J, Boag S, Stewart J R, Frost C D, Skoda M W A, Parnell S R and Babcock E 2011 *Physica B* **406** 2436
[28] Romalis M V and Cates G D 1998 *Phys. Rev. A* **58** 3004
[29] Qin Z C, Huang C Y, Buck Z N, Kreuzpaintner W, Amir S M, Salman A, Ye F, Zhang J P, Jiang C Y, Wang T and Tong X 2021 *Chin. Phys. Lett.* **38** 052801
[30] Batz M, Baeßler S, Heil W, Otten E W, Rudersdorf D, Schmiedeskamp J, Sobolev Y and Wolf M 2005 *J. Res. Natl. Inst. Stand. Technol.* **110** 293
[31] Babcock E, Nelson I A, Kadlecek S and Walker T G 2005 *Phys. Rev. A* **71** 013414