Numerical simulation of wall relaxation in Rb vapor optically pumped with $^{21}$Ne

D A Dolgopyatov, E S Navasardyan and I A Arkharov
Bauman Moscow State Technical University, 2nd Baumanskaya St., 5, Moscow, 105005, Russia
E-mail: arkharov@bmstu.ru

Abstract. Spatial distribution of the polarization in Rb vapor optically pumped with $^{21}$Ne has been modeled numerically with 40 mW laser beam power, 180°C cell temperature, 1 bar cell pressure, and 6 mm $\times$ 15 mm pancake-cell geometry. An inhomogeneity of polarization throughout the cell results from a wall relaxation of polarized media and a decay of the pumping beam. For small cells characteristic of experimental conditions, a contribution of wall relaxation to the total alkali relaxation rate can become significant, demanding more accurate evaluation than afforded by analytical model based on average parameters.

1. Introduction
Isotopes of noble gases with non-zero nuclear spin—$^3$He, $^{21}$Ne, $^{83}$Kr, $^{129}$Xe, $^{131}$Xe—can be utilized for nuclear magnetic resonance (NMR) imaging in medical or scientific applications. This work on $^{21}$Ne has been expedited by a $^3$He supply crisis, opening the way to alternative biomarkers, as well as by the progress in the cryogenic technology of isotope enrichment [1].

To increase signal-to-noise ratio, the nuclear spins of isotopes should be hyperpolarized well above thermal equilibrium under conditions of tomographic magnetic field strength. This is performed with spin-exchange optical pumping (SEOP) of noble gas isotopes in the presence of alkali vapor [2].

In the present paper, we numerically simulate the steady-state polarization of Rb vapor, optically pumped with $^{21}$Ne buffer gas, accounting for its 2D spatial distribution caused by wall relaxation, and compare the results with an analytical solution based on a wall relaxation rate averaged throughout the cell. The media properties for the simulation were taken from [3]–[6].

2. Theory
SEOP is a process of angular momentum transfer from circularly polarized light to alkali (Rb) electron polarization of Zeeman sub-levels and via hyperfine interactions to noble gas ($^{21}$Ne) nuclear polarization of Zeeman sub-levels. The alkali nuclei also become polarized. These traits can be modeled with the following equations [6]–[7].

Equilibrium alkali polarization equation:

$$ P^a = \frac{1}{\epsilon^a+1} \cdot \frac{R_{op}}{R_{op} + R_{rel}^a} $$

(1)
Pumping beam decay equation:

\[ \frac{dR_{op}}{dz} = -n^a \sigma(\omega)(1 - P^a)R_{op} \]  

(2)

Equilibrium noble gas polarization equation:

\[ P^b = P^a \frac{\epsilon^b}{R^b/\epsilon^b} \cdot \frac{R^b_{ba}}{R^b_{ba} + 1/T^b} \]  

(3)

Their parameters are further given by:

\[ R_{rel} = \frac{1}{\epsilon^a + 1} (R_{sd}^a + R_{sd}^b + R_{se}^b + R_{pr}) + R_{wall}^a \]  

(4)

\[ R_{sd}^a = n^a \sigma_{sd}^a \nu^a \]  

(5)

\[ R_{bd}^b = n^b \sigma_{sd}^b \nu^b \]  

(6)

\[ v^{ij} = \left( \frac{8kT}{\pi M} \right)^{1/2} \]  

(7)

\[ M = \left( \frac{1}{m^i} + \frac{1}{m^j} \right)^{-1} \]  

(8)

\[ R_{ba}^b = R_{ba}^b \frac{n^b}{\epsilon^a} \]  

(9)

\[ R_{se}^b = \kappa_{se} n^a \]  

(10)

\[ R_{wall}^a = D^a \left( \frac{\pi}{r} \right)^2 \]  

(11)

\[ \epsilon(I, \beta) = (2I + 1) \text{cth}(\beta/2) \text{cth}[\beta(I + 1/2)] - \text{cth}^2(\beta/2) \]  

(12)

\[ \beta = \frac{1 + P}{1 - P} \]  

(13)

Here, superscripts \( a \) and \( b \) refer to alkali and noble gas, respectively; \( i \) and \( j \) to species of corresponding type; \( P \) is polarization; \( n \) is atomic density, \( \text{m}^{-3} \); \( R_{op} \) is the optical pumping rate, \( \text{s}^{-1} \); \( R_{rel} \) is the relaxation rate, \( \text{s}^{-1} \); \( R_{sd} \) is the spin destruction rate, \( \text{s}^{-1} \); \( R_{se} \) is the spin exchange rate, \( \text{s}^{-1} \); \( R_{pr} \) is the probe-beam pumping rate, \( \text{s}^{-1} \); \( R_{wall} \) is the wall relaxation rate, \( \text{s}^{-1} \); \( T \) is a longitudinal relaxation time constant, \( \text{s} \); \( z \) is a coordinate along pumping beam axis, \( \text{m} \); \( \sigma \) is the photon-absorption cross section, \( \text{m}^2 \); \( \omega \) is the pumping beam frequency, Hz; \( \sigma_{sd} \) is the spin-destruction cross section, \( \text{m}^2 \); \( \nu \) is the relative velocity of colliding particles, \( \text{m/s} \); \( k \) is the Boltzmann constant; \( T \) is cell temperature, K; \( M \) is the reduced mass of colliding particles, \( \text{kg} \); \( m \) is the mass of a particle, \( \text{kg} \); \( D \) is the diffusion coefficient, \( \text{m}^2/\text{s} \); \( I \) is nuclear spin; \( S \) is electron spin; \( \epsilon \) is a paramagnetic coefficient; \( \beta \) is spin temperature, \( \text{r} \) is the spherical cell radius, \( \text{m} \); and \( \kappa_{se} \) is the spin-exchange-rate coefficient, \( \text{m}^3/\text{s} \).

This model neglects the relaxation due to magnetic-field inhomogeneity of the cell and the presence of quenching nitrogen, \( \text{N}_2 \). The probe beam is not considered further in this paper, thus \( R_{pr} = 0 \).

The model above has the following convenient analytical solution, which permits the calculation of 1D polarization distributions of Rb and \( ^{21}\text{Ne} \) using equations (1) and (3):

\[ R_{op}(z) = (\epsilon^a + 1)R_{rel}^a \cdot W \left[ \frac{1}{\epsilon^a + 1} \cdot \exp \left[ \frac{\lambda_{D1}^a R_{op-int}^a}{R_{rel}^a} \right] \right] \]  

(14)
\[ R_{\text{op-int}} = \frac{\sigma(\lambda D_1) P_{\text{Pump}} \lambda D_1}{Ahc} \]

where \( W \) is a Lambert W-function; \( \lambda D_1 \) is the Rb D1-transition wavelength, m; \( P_{\text{Pump}} \) is pumping-beam power, W; \( A \) is the cell cross-sectional area, \( m^2 \); \( h \) is Planck’s constant; and \( c \) is the speed of light.

One can evaluate the normalized contribution of the wall relaxation rate \( R_{\text{wall}}^a \) for different temperatures, pressures, and cell radii. Figure 1 shows a sample of such calculations. The normalized contribution of wall relaxation is significant for low buffer-gas pressure and small cell size, which can be the case in laboratory experiments. For example, under the conditions simulated, the \( R_{\text{wall}}^a \) is as high as 34.3% the total alkali relaxation rate, assuming an equivalent cell radius

\[ r = \frac{3V}{A} \]

where \( V \) is cell volume, \( m^3 \).

**Figure 1.** Normalized wall relaxation rate for spherical cells with radii of 2 to 25 mm.

The evaluation of \( R_{\text{wall}}^a \) by equation (14) provides a rough estimate of its effective average value, which is even less precise for non-spherical cells. To account for wall relaxation more accurately, equation (1) should be substituted by the alkali diffusion equation:

\[ D^a \nabla^2 P^a + \frac{2Q^a}{n^a} = 0 \]

\[ Q^a = n^a \left( \frac{1}{e^a + 1} R_{\text{op}}^a(1 - P^a) - R_{\text{rel}}^a P^a \right) \]

\[ R_{\text{rel}}^a = \frac{1}{e^a + 1} (R_{sd}^{sa} + R_{sd}^{ab} + R_{se}^{ab} + R_{pr}) \]
Figure 2. Results of numerical simulation: Rb polarization and normalized pumping rate vs. z-coordinate.

where $Q$ is the intensity of polarized particle sources, $m^{-3}s^{-1}$. Equation (17) has been solved numerically with the following boundary conditions:

$$D\frac{\partial P_a(r, z)}{\partial z} \bigg|_{z=0} = \alpha_a P_a(r, 0) \quad (20)$$

$$D\frac{\partial P_a(r, z)}{\partial z} \bigg|_{z=L} = -\alpha_a P_a(r, L) \quad (21)$$

$$\frac{\partial P_a(r, z)}{\partial r} \bigg|_{r=0} = 0 \quad (22)$$

$$D\frac{\partial P_a(r, z)}{\partial r} \bigg|_{r=R} = -\alpha_a P_a(R, z) \quad (23)$$

$$\alpha = \frac{1}{4} \left( \frac{8kT}{\pi m} \right)^{1/2} \quad (24)$$

where $r$ is the cell radial coordinate, $m$; and $L$ is the cell length, $m$.

Equations (20) and (21) represent the wall relaxation at cell edges, equation (22) – a condition of symmetry on the cell axis, and equation (23) – wall relaxation on the cell’s outer surface. Equation (24) was derived from the average wall collision frequency per particle, assuming a Maxwell distribution. Equation (2) was used for pump beam intensity calculations.

3. Results

Figure 2 presents the results of numerical simulation of Rb vapor polarization under 40 mW laser beam power, 180°C cell temperature, 1 bar cell pressure, 6 mm × 15 mm pancake-cell geometry.

The average simulated polarization of Rb across the cell is 91.3%, versus a value of 99.4% predicted by the analytical formula. These correspond to cell-average $^{21}$Ne polarizations of 7.5% and 8.2%, respectively. 1D numerical simulation, neglecting the relaxation on the cell’s outer surface, gives 98% polarization of Rb and 8.1% polarization of $^{21}$Ne.
4. Conclusion and future work

The simulated Rb and $^{21}\text{Ne}$ polarizations are in fair agreement with those predicted analytically using cell-average values for the wall relaxation rate $R_{\text{wall}}$. The error reaches 8.9% and contains a significant contribution of the relaxation on the cell’s outer surface. 1D numerical simulation gives 1.4% difference with analytical solution and includes the wall relaxation at cell edges only. Minor corrections are possible in terms of accounting for neglected effects. However, it is recommended that numerical simulations should be performed for laboratory-like conditions, especially in cases with small cells and low buffer-gas pressures. The contribution of $R_{\text{wall}}$ vanishes with increasing cell size.

The numerical simulation does exhibit differing qualitative results compared with the analytical formula. The boundary layers feature a marked decrease of polarization with a corresponding pumping-light absorption. This is especially important for the front edge of the cell, where the pumping beam’s power can be completely diminished under certain conditions. Within the cell core, however, the polarization is fairly uniform.

Future work will include extensive simulation of $^{21}\text{Ne}$ polarization with K, Rb, and Cs under different cell conditions and with varying noble-gas isotope enrichments and modeling of transient effects.

References

[1] Arkharov A M, Arkharov I A, Dolgopyatov D A, and Bondarenko V L 2013 $^3\text{He}$ supply crisis: reasons and challenges Chemical and Petroleum Engineering 49 41–45
[2] Conradi M S, Saam B T, Yablonskiy D A and Woods J C 2006 Hyperpolarized $^3\text{He}$ and perfluorocarbon gas diffusion MRI of lungs Progress in Nuclear Magnetic Resonance Spectroscopy 48 63–83
[3] Alcock C B, Itkin V P and Horrigan M K 1984 Canadian Metallurgical Quarterly 23 309
[4] Allred J C, Lyman R N, Kornack T W and Romalis M V 2002 High-Sensitivity Atomic Magnetometer Unaffected by Spin-Exchange Relaxation Phys. Rev. Lett. 89 130801
[5] Franz F A and Volk C 1976 Spin relaxation of rubidium atoms in sudden and quasimolecular collisions with light-noble-gas atoms Phys. Rev. A 14 1711–28
[6] Ghosh R J and Romalis M V 2010 Measurement of spin-exchange and relaxation parameters for polarizing $^{21}\text{Ne}$ with K and Rb Phys. Rev. A 81 043515
[7] Walker T G and Happer W 1997 Spin-exchange optical pumping of noble gas nuclei Reviews of Modern Physics 69 629–642