

Some Consequences of the Presence of N₂ Gas in Polarized ³He Target Cells

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Abstract

A small amount of N₂ gas is needed to increase the efficiency of optical pumping by suppressing radiation trapping. In addition, the intensity of the D2 fluorescence and the rate of beam induced relaxation also depend on the N₂ density.

1 Introduction

N₂ gas in a polarized ³He target cell plays a role in at least four different ways (in order of importance):

1. Improves the efficiency of optical pumping by suppressing radiation trapping
2. Increases the dilution factor in electron scattering
3. Decreases the beam induced relaxation of ³He in the target chamber
4. Increases the spin relaxation of Rb (and possible ³He?) in the cell
5. Has a relationship to the intensity of the D2 fluorescence used in EPR ³He polarimetry

We will describe the mechanism of radiation trapping and give an estimate for the fraction of excited atoms that decay radiatively. We will calculate and discuss the [N₂] dependence of the beam induced relaxation rate. We will not discuss the effect of N₂ on the dilution factor. This must be evaluated carefully on an experiment by experiment basis and is beyond the scope of this document. We will derive a relationship between the intensity of D2 fluorescence and density of N₂. Finally we will take a careful look at frequently used guideline of filling the cell with about 70 torr of N₂.

2 Radiation Trapping

Radiation trapping occurs when a photon is emitted by one atom and reabsorbed by another atom. Its effect on the efficiency of optical pumping can be understood qualitatively as follows. Consider a vapor of a fictional alkali atom with no nuclear spin in a magnetic field. (The case for an alkali atom with nuclear spin is qualitatively similar.) A beam of *right* circularly polarized light propagating in the direction of the magnetic field tuned to the D1 transition will excite the atom from the $S_{1/2}$, $m = -1/2$ ground state to the $P_{1/2}$, $m = +1/2$ excited state. In the absence of nonradiative quenching, the excited state will decay spontaneously with the emission of a photon. (For the laser intensities typically used in optical pumping, the spontaneous decay rate greatly exceeds the stimulated emission rate). In the absence of ³He, this emitted photon has a 2/3 chance of being *left* circularly polarized and a 1/3 chance of being *linearly* polarized. However, under our conditions, the presence of on order of 10 amg of ³He results in complete depolarization of the excited atoms. If the vapor is dense and the average path length out of the cell is long, then this photon will likely be reabsorbed by another atom in the cell. Because this photon is on average unpolarized,

parameter	description	$P_{1/2}$	$P_{3/2}$	units	source
τ	natural lifetime	27.70	26.35	ns	NIST
τ^{-1}	spontaneous decay rate	36.10	37.95	MHz	NIST
σ_q	N ₂ quenching cross section	58	42.5	10 ⁻²⁰ m ²	happer
k_q	N ₂ quenching rate constant	11	8.1	GHz/amg	happer
σ_m	N ₂ mixing cross section	15	23.5	10 ⁻²⁰ m ²	happer
k_m	N ₂ mixing rate constant	2.9	4.5	GHz/amg	happer
σ_d	He depolarization cross section	9	100	10 ⁻²⁰ m ²	happer
k_d	³ He depolarization rate constant	4.6	51	GHz/amg	happer

Table 1: Quenching and Mixing Parameters for Rb-N₂ Collisions.

any subsequent reabsorption usually results in the depolarization of the absorbing atom. After the photon is reabsorbed, the newly excited atom can again spontaneously decay by emitting a photon. In principle, the same photon can undergo many cycles of reabsorption and emission and therefore becomes “trapped” in the cell by the vapor. Ultimately the trapped radiation behaves as an additional relaxation mechanism to the alkali vapor. Estimating the quantitative effect of radiation trapping is difficult because it depends on the intensity of the pump beam, the geometry of the cell, and the polarization & density of the alkali vapor.

3 Nonradiative Quenching

The effect of radiation trapping can be suppressed if the excited atoms are preferentially nonradiatively “quenched.” This process occurs when the the excited atoms return to the ground state by collisionally transferring the excitation energy to a buffer gas particle. Molecular gases such a N₂ have quenching cross sections that are at least three orders of magnitude larger than inert gases such as He. Collisions with molecular gases provide “more ways” (or greater phase space) for energy conservation than collisions with inert gases because molecular gases possess rotational and vibrational degrees of freedom. Although the cross section is at least 3 orders of magnitude smaller for He quenching, the density of He is 2 orders of magntiude larger than the N₂ density. Therefore He contributes at most 10 percent of the quenching rate and will be ingored in the following calculations.

The quenching rate is given as:

$$\Gamma_q = [\text{N}_2]\sigma_q \langle v \rangle_{\text{rel}} = k_q[\text{N}_2] \sqrt{\frac{T}{500 \text{ K}}} \quad (1)$$

where σ_q is the quenching cross section and $\langle v \rangle_{\text{rel}}$ is the mean relative thermal velocity between the Rb and N₂:

$$\langle v \rangle_{\text{rel}} = \sqrt{\frac{8kT}{\pi} \left[\frac{1}{m_{\text{N}_2}} + \frac{1}{m_{\text{Rb}}} \right]} \quad (2)$$

The quenching rate constant k_q is obtained under the assumption that the cross section is independent of temperature. By noting that the spontaneous decay rate is the inverse of the excited state lifetime, τ , we find that the fraction of Rb atoms that decay radiatively is:

$$f_r = \frac{\tau^{-1}}{\tau^{-1} + \gamma_q} = \frac{1}{1 + \tau\gamma_q} \quad (3)$$

Under typical operating conditions, $[\text{N}_2] \approx 0.08$ amg and $T \approx 500$ K, the fraction of Rb atoms that decay radiatively is ≈ 0.05 or alternatively $\approx 5\%$.

plot of n2 density vs fraction of rad decays at 230 , 260 , and 290 C

4 Beam Induced ^3He Relaxation

Ionizing radiation increases the nuclear spin relaxation in the target chamber. Also known as “beam depolarization,” it is essentially a two step process. First, the beam ionizes an ^3He atom which results in a free electron and an atomic ion $^3\text{He}^+$. There is also the possibility that the atomic ion bonds with a neutral ^3He atom to form a molecular ion $^3\text{He}_2^+$. Second, interactions with ^3He ions induce ^3He nuclear spin flips. Therefore, the total relaxation rate due to ionization by the beam is given by:

$$\Gamma_{\text{ion}} = \left(\frac{1}{20 \text{ hrs}}\right) \cdot \left(\frac{I}{10 \mu\text{A}}\right) \cdot \left(\frac{2.0 \text{ cm}^2}{A_{\text{tc}}}\right) \cdot (n_a + n_m) \quad (4)$$

where I is the electron beam current, A_{tc} is the mean cross sectional area of the target chamber, and n_a & n_m are the average number of spins lost per atomic ion created due to interactions with atomic & molecular ions respectively.

Atomic ions contribute to polarization loss due to a “spin-exchange”-like interaction between the ^3He nucleus and the unpaired electron in the atomic ion. Because charge exchange occurs readily, electrons from highly polarized neutral atoms jump to lowly polarized atomic ions. The newly formed atomic ion partially depolarizes until it undergoes charge exchange and so on. The cumulative effect is at most one nuclear spin flip [?]. In addition to this process, molecular ions also lose polarization to the rotational degrees of freedom via a spin-rotation interaction [?]. Little mention is made in the literature about relaxation due to interactions with free electrons, so we shall ignore them as well.

The presence of a foreign gas such as N_2 limits the lifetime of ^3He ions and consequently limits the beam induced relaxation rate. Molecular ions have the greater potential to depolarize many nuclei, but their lifetimes are greatly shortened by N_2 . Consequently we can safely set $n_m \approx 0$. This is not true for atomic ions and it is quite tedious to calculate n_a directly using the formalism of blah. A useful “homemade” parameterization can be given in matrix form:

$$n_a(h, \rho) = \begin{bmatrix} 1 \\ h \\ h^2 \\ h^3 \end{bmatrix}^T \begin{bmatrix} +1.0283\text{E}+0 & +2.2063\text{E}-1 & -5.4940\text{E}-2 & +4.9159\text{E}-3 \\ +1.9515\text{E}-1 & -9.0280\text{E}-1 & +1.6384\text{E}-1 & -1.2999\text{E}-2 \\ -9.8146\text{E}-1 & +7.6918\text{E}-1 & -1.1821\text{E}-1 & +8.0086\text{E}-3 \\ +3.5643\text{E}-1 & -1.9663\text{E}-1 & +2.5395\text{E}-2 & -1.2960\text{E}-3 \end{bmatrix} \begin{bmatrix} 1 \\ \rho \\ \rho^2 \\ \rho^3 \end{bmatrix} \quad (5)$$

where:

$$h = \frac{[^3\text{He}]_{\text{tc}}}{10 \text{ amg}} \quad \& \quad \rho = 100 \cdot \frac{[\text{N}_2]_{\text{tc}}}{[^3\text{He}]_{\text{tc}}} \quad (6)$$

This parameterization reproduces the full calculation of n_a to better than 0.5% for ^3He densities from 7 to 14 amg ($h = [^3\text{He}]_{\text{tc}}/10 \text{ amg}$) with N_2 to ^3He density ratios ($\rho \equiv 100 \cdot [\text{N}_2]_{\text{tc}}/[^3\text{He}]_{\text{tc}}$) from 1/3% to 3%. If the ^3He density is 11 amg or $h = 1.1$, the the matrix collapses to give:

$$n_a(h = 1.1, \rho) = 0.59842 - 0.10962 \cdot \rho + 0.016085 \cdot \rho^2 - 0.0013705 \cdot \rho^3 \text{ REDO} \quad (7)$$

plot na

5 Intensity of D2 Fluorescence

The presence of N_2 results in very efficient mixing between the $P_{1/2}$ and $P_{3/2}$ excited states. Thus when the excited atoms do decay radiatively, they undergo both the D1 and D2 transitions with roughly equal probabilities. This implies the presence of D2 fluorescence. The intensity of this light has the following form:

$$\begin{aligned} I_{D2} = & \text{(complicated geometrical/density/polarization factor)} \\ & \times \text{(fraction of decays that are radiative)} \\ & \times \text{(fraction of excited atoms in the } P_{3/2} \text{ state)} \end{aligned} \quad (8)$$

Because the mixing and quenching rates are of the same order of magnitude, one must solve the full rate equations to find the fraction of excited atoms in the $P_{3/2}$ state:

$$\dot{s} = -s(\gamma_{\text{op1}} + \gamma_{\text{op2}}) + p_1(\Gamma_{q1} + 1/\tau_1) + p_2(\Gamma_{q2} + 1/\tau_2) \quad \text{wrong include polarization and stimulated emmission} \quad (9)$$

$$\dot{p}_1 = +s\gamma_{\text{op1}} - p_1(\Gamma_{q1} + \Gamma_{m1} + 1/\tau_1) + p_2\Gamma_{m2} \quad (10)$$

$$\dot{p}_2 = +s\gamma_{\text{op2}} - p_2(\Gamma_{q2} + \Gamma_{m2} + 1/\tau_2) + p_1\Gamma_{m1} \quad (11)$$

where s , p_1 , & p_2 are respectively the populations of the $S_{1/2}$ ground state, $P_{1/2}$ excited state, & $P_{3/2}$ excited state, $\gamma_{\text{op1(2)}}$ is the optical pumping rate to the $P_{1/2}$ ($P_{3/2}$) state, and $\Gamma_{m1(2)}$ is the collisional mixing rate from the $P_{1/2}$ ($P_{3/2}$) excited state to the $P_{3/2}$ ($P_{1/2}$) excited state:

$$\Gamma_m = [N_2]\sigma_m \langle v \rangle_{\text{rel}} = k_m[N_2] \sqrt{\frac{T}{500 \text{ K}}} \quad (12)$$

Setting all the rates to zero gives the equilibrium populations of the excited state relative to the ground state:

$$p_1/s = \frac{\gamma_{\text{op1}} + \gamma_{\text{op2}} \left[\frac{\Gamma_{m2}}{\Gamma_{q2} + 1/\tau_2 + \Gamma_{m2}} \right]}{\Gamma_{q1} + 1/\tau_1 + \Gamma_{m1} \left[\frac{\Gamma_{q2} + 1/\tau_2}{\Gamma_{q2} + 1/\tau_2 + \Gamma_{m2}} \right]} \quad (13)$$

$$p_2/s = \frac{\gamma_{\text{op2}} + \gamma_{\text{op1}} \left[\frac{\Gamma_{m1}}{\Gamma_{q1} + 1/\tau_1 + \Gamma_{m1}} \right]}{\Gamma_{q2} + 1/\tau_2 + \Gamma_{m2} \left[\frac{\Gamma_{q1} + 1/\tau_1}{\Gamma_{q1} + 1/\tau_1 + \Gamma_{m1}} \right]} \quad (14)$$

By imposing the constraint $s + p_1 + p_2 = 1$, we get fraction of atoms in the ground state:

$$s = \left(1 + \frac{\gamma_{\text{op1}} + \gamma_{\text{op2}} \left[\frac{\Gamma_{m2}}{\Gamma_{q2} + 1/\tau_2 + \Gamma_{m2}} \right]}{\Gamma_{q1} + 1/\tau_1 + \Gamma_{m1} \left[\frac{\Gamma_{q2} + 1/\tau_2}{\Gamma_{q2} + 1/\tau_2 + \Gamma_{m2}} \right]} + \frac{\gamma_{\text{op2}} + \gamma_{\text{op1}} \left[\frac{\Gamma_{m1}}{\Gamma_{q1} + 1/\tau_1 + \Gamma_{m1}} \right]}{\Gamma_{q2} + 1/\tau_2 + \Gamma_{m2} \left[\frac{\Gamma_{q1} + 1/\tau_1}{\Gamma_{q1} + 1/\tau_1 + \Gamma_{m1}} \right]} \right)^{-1} \quad (15)$$

Although in principle we should combine Eqn. (9) with Eqns. (10) & (11) to solve for p_1 and p_2 , under our conditions, it is safe to assume $s \approx 1$. Assuming that we are only pumping to the D1 transition ($\gamma_{\text{op2}} = 0$), we find:

$$I_{D2} \propto \left(\frac{1}{1 + \tau_2 \Gamma_{q2}} \right) \left(\frac{\gamma_{\text{op1}}}{\Gamma_{q2} + 1/\tau_2 + \Gamma_{m2} \left[\frac{\Gamma_{q1} + 1/\tau_1}{\Gamma_{q1} + 1/\tau_1 + \Gamma_{m1}} \right]} \right) \left(\frac{\Gamma_{m1}}{\Gamma_{q1} + 1/\tau_1 + \Gamma_{m1}} \right) \quad (16)$$

In the limit that the N_2 density approaches zero, the D2 fluorescence also approaches zero because the collisional mixing rate also approaches zero. In the limit that the N_2 density approaches infinity, the D2 fluorescence approaches zero again because now the nonradiative quenching rate approaches infinity. This seems to imply that there is an ideal N_2 density that maximizes the D2 fluorescence. **plot of n2 density vs fraction of rad decays at 230 , 260 , and 290 C**

6 How much N_2 should we put in the cell?

P_{goal} at 23°C torr	PC operating amagats	TC operating amagats	notes
60 to 150	0.073 to 0.18		one chamber 1987 chupp
75	0.091		one chamber amy barton
100	0.099	0.14	two chamber 1992 chuppp
30 to 100	0.036 to 0.12		one chamber 1994 chupp
65	0.066	0.087	middleton E142
60	0.064	0.087	romalis E154
60 to 70	0.079	0.12	kominis GDH
65	0.078	0.12	kramer g2n
70	0.075	0.11	saGDH
90	0.095	0.17	LPC-GE _n

Table 2: Fill Pressure of N₂. **references!!!!**