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# High nuclear polarization of helium-3 at low and high pressure by metastability exchange optical pumping at 1.5 Tesla

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**Abstract:** We perform metastability exchange optical pumping of helium-3 in a strong magnetic field of 1.5 T. The achieved nuclear polarizations, from 80% at 1.33 mbar to 25% at 67 mbar, show a substantial improvement at high pressures with respect to standard low-field optical pumping. The specific mechanisms of metastability exchange optical pumping at high field are investigated, advantages and intrinsic limitations are discussed. From a practical point of view, our results open the way to alternative technological solutions for polarized helium-3 applications and in particular for magnetic resonance imaging of human lungs.

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A gas of ground state  $^3\text{He}$  atoms in which a high degree of nuclear polarization is achieved offers an incredibly rich playground in various fields of science from statistical or nuclear physics to biophysics and medicine [1]. Depending on the targeted application, the degree of nuclear polarization, the sample density or the production rate of polarized atoms should be optimized. A recent application which may have an important impact in the diagnosis of pulmonary diseases is polarized gas magnetic resonance imaging (MRI) [2]. Clinical studies to demonstrate the relevance of this new tool are under way in the United States and in Europe. Yet, if a wide expertise exists in MRI to adapt the existing imaging techniques to the case of polarized gases, the gas preparation remains a critical stage to be transferred from physics laboratories to hospitals. Two methods are presently used to polarize  $^3\text{He}$ : spin-exchange with optically pumped alkali atoms [3] and pure-He metastability exchange optical pumping (MEOP) [4]. In standard conditions, MEOP is performed at low pressures (1 mbar) in a guiding magnetic field of the order of 1 mT. Circularly polarized light at 1083 nm corresponding to the  $2^3\text{S}-2^3\text{P}$  transition of He is used to transfer angular momentum to the atoms and nuclear polarization is created by hyperfine coupling in the metastable  $2^3\text{S}$  state. Through metastability exchange collisions nuclear polarization builds up in the ground state  $^3\text{He}$  atoms. The efficiency of MEOP in standard conditions rapidly decreases if the pressure of the sample exceeds a few mbar (insert in Fig.6-a) [5, 6, 7]. Therefore a delicate polarization-preserving compression stage is necessary for MRI in which the gas should be at atmospheric pressure and for all applications needing a dense sample. In this letter, we show that a strong magnetic field of 1.5 T improves the efficiency of MEOP with respect to standard low-field optical pumping. At 1 mbar, we easily obtain 80% of nuclear polarization with much lighter experimental constraints. At higher pressures, the achieved nuclear polarizations are

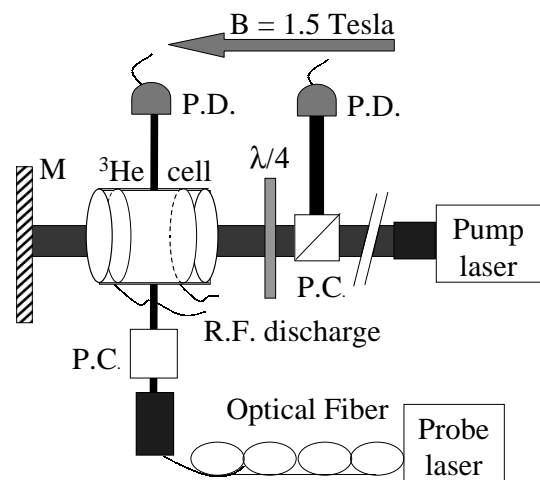


FIG. 1: Experimental setup. P.D.: photodiode, P.C.: polarizing beam-splitter,  $\lambda/4$ : quarter wave plate, M: mirror.

dramatically increased. This indicates that a relaxation channel which severely limits the efficiency of MEOP at high pressure is inhibited at high field. Optical pumping at high magnetic field and high pressure could thus constitute an attractive option for applications.

Experiments are performed in the bore of the 1.5 T superconducting magnet of a clinical MRI system. The experimental apparatus is sketched in Fig.1. The helium gas is enclosed in a sealed cylindrical Pyrex cell (diameter 5 cm, length 5 cm). Four cells filled with 1.33 mbar, 8 mbar, 32 mbar and 67 mbar of pure  $^3\text{He}$  are used. A radio-frequency discharge at 3 MHz sustained by external electrodes is used to maintain the metastable atoms density  $n_m$  in the range  $10^{10}-10^{11}$  atoms/cm<sup>3</sup>, depending on the discharge intensity, at all pressures. The optical pumping laser is either a 50 mW single mode laser diode amplified to 0.5 W or a 2 W broadband fiber laser (1.63 GHz FWHM). The slightly divergent pump beam

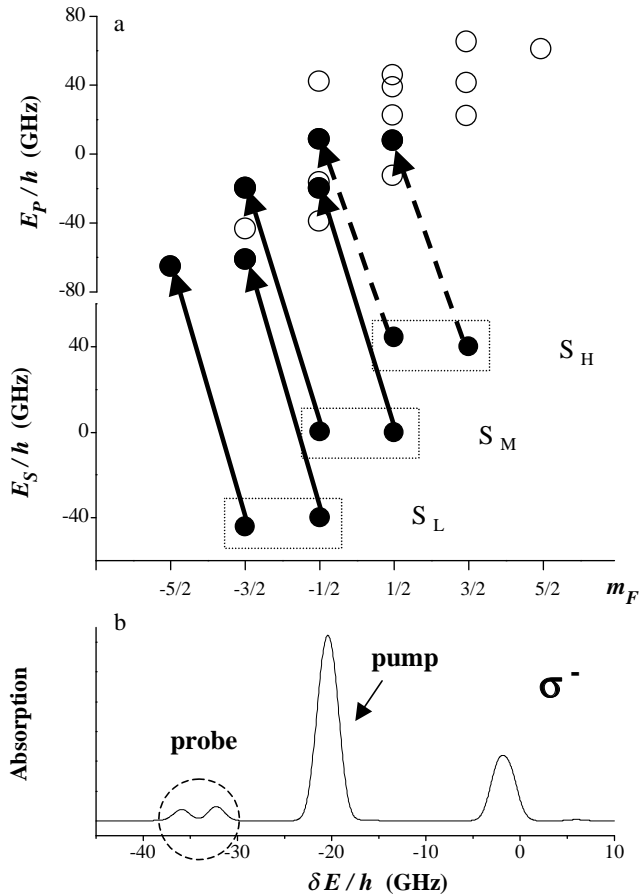


FIG. 2: a- Energy level scheme of  ${}^3\text{He}$  at 1.5 T for the metastable  $2^3\text{S}$  state  $E_S$  and the  $2^3\text{P}$  state  $E_P$ .  $\sigma^-$ -polarized pump (full arrows) and probe (dashed arrows) transitions. Energy zeroes in the  $2^3\text{S}$  and  $2^3\text{P}$  states are those of the sublevels involved in the  ${}^4\text{He-D}_2$  line at  $B=0$ , also used as the origin for the energy of the transitions. b- Complete absorption spectrum for  $\sigma^-$  light.

reaches a diameter of about 1 cm FWHM in the cell. It is back-reflected to enhance its absorption and collected by a photodiode to monitor its tuning to atomic resonance. A single mode laser diode is used as a weak probe beam to measure the nuclear polarization. It is linearly polarized perpendicularly to the magnetic field ( $\sigma$  polarization). The discharge intensity is modulated at 133 Hz and the probe absorption is measured with a lock-in amplifier. Laser sources and all the electronics remain several meters away from the magnet bore in a low-field region.

At 1.5 T the energy levels of the  $2^3\text{S}$  and  $2^3\text{P}$  states are spread over 80 and 160 GHz respectively due to Zeeman splitting (Fig.2). We note  $m_J$ ,  $m_I$ ,  $m_F$  the magnetic quantum numbers for the electronic, nuclear, and total angular momentum. In the  $2^3\text{S}$  state the sublevels are arranged into three pairs of quasi-degenerate levels of increasing energy ( $S_L$ ,  $S_M$ , and  $S_H$ ), that correspond respectively to  $m_J=-1$ , 0, and 1 in the completely de-

coupled limit  $B \rightarrow \infty$ . In fact at 1.5 T hyperfine decoupling in the  $2^3\text{S}$  state is already important so that the eigenstates of the Hamiltonian show only little mixing of the order of  $\epsilon=1\%$  between different eigenstates  $|m_J, m_I\rangle$  of the decoupled system. Optical pumping was found to be the most efficient with a  $\sigma^-$ -polarized pump laser tuned to simultaneously depopulate four metastable sublevels (Fig.2). These transitions have a total oscillator strength close to 4, which guarantees strong pump laser absorption in contrast with the low-field situation where the most efficient ( $C_8$  and  $C_9$ ) optical pumping transitions have a weak oscillator strength (0.29 and 0.37 respectively). The atoms are pumped into the pair  $S_H$  by the following sequence: laser excitation, collisional redistribution in the  $2^3\text{P}$  state and spontaneous emission. The ground state nuclear polarization  $M$  is defined as  $M=(n_+ - n_-)/(n_+ + n_-)$  where  $n_+$  and  $n_-$  denote populations of the  $m_I=1/2$  and  $m_I=-1/2$  nuclear spin states respectively. In the absence of optical pumping, metastability exchange collisions impose a spin temperature distribution for the  $2^3\text{S}$  sublevel populations, proportional to  $e^{\beta m_F}$  where  $M=(e^\beta - 1)/(e^\beta + 1)$ . To measure  $M$ , we probe the populations of the two  $m_F=1/2$  and  $m_F=3/2$  states that are not addressed by the pump. By sweeping the probe laser frequency, the corresponding spectra are recorded both for  $M=0$  and for optically-pumped steady-state situations. The spin temperature and the nuclear polarization are inferred from the relative heights of the peaks before and after optical pumping (Fig.3-a). By tuning the probe laser frequency on the  $m_F=3/2$  line, we can monitor the build-up of the polarization, as well as its decay when the pump is turned off (Fig.3-b). These measurement procedures operate in arbitrary magnetic field and pressure [8].

In Fig.4, we present results on the steady-state polarization  $M$  and on the polarization build-up time constant  $t_b$  in the 1.33 mbar cell as a function of the discharge intensity characterized by the polarization decay time  $T_1$ . We distinguish two regimes: weak and strong discharges. In weak discharges ( $T_1 \geq 300$  s), the achieved polarization with the broadband 2 W laser is high, about 80% independently of  $T_1$ . The polarization build-up is however quite slow ( $t_b$  ranging from 60 to 350 s). In strong discharges, build-up is much faster ( $t_b$  ranging from 15 to 55 s) although the steady-state polarization is lower. Fig.5 shows the influence of the pump laser power  $\mathcal{P}_L$  for a given discharge ( $T_1 = 300$  s). A power as low as 0.5 W is sufficient for the polarization and the build up time to almost reach their asymptotic values. Signal to noise ratio is better than  $10^3$  and the estimated error on the steady-state polarization is  $\leq 2\%$ . Yet, scatter of results is of the order of  $\pm 10\%$  over several runs for the same parameters. This is attributed to plasma conditions that are difficult to exactly reproduce in high field. Similar studies of optical pumping dynamics have been performed at higher pressures. Selected results for one

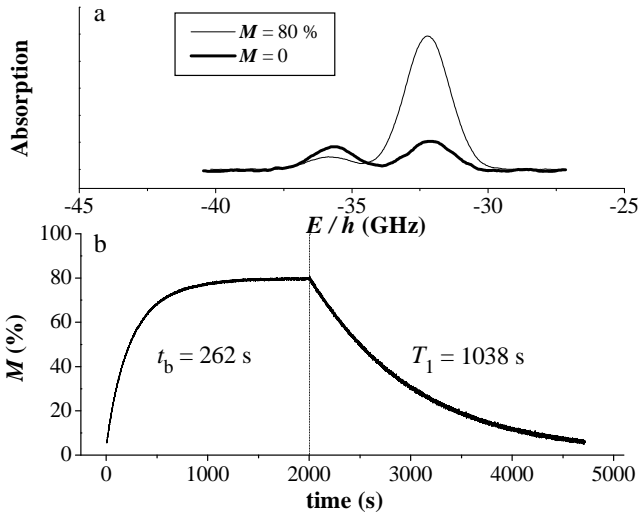


FIG. 3: Example of signal in the 1.33 mbar cell: a- Optical measurement of polarization and b- Polarization build-up and decay. Optical pumping laser is applied at time  $t=0$ , turned off after 2000 s.

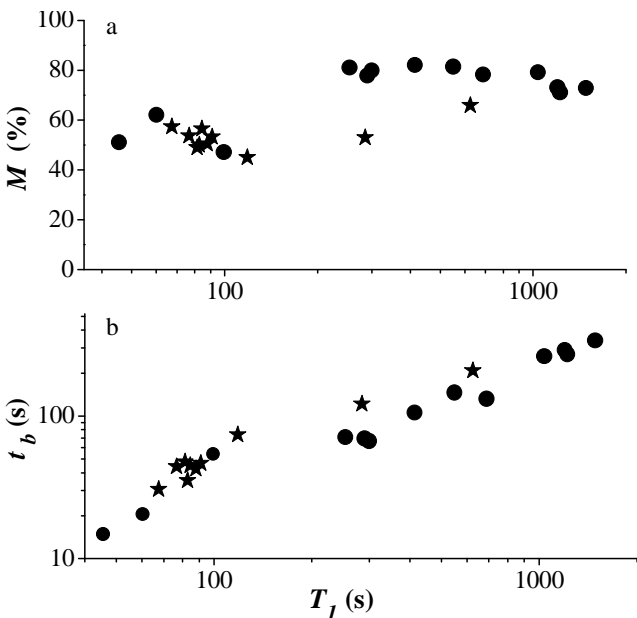


FIG. 4: 1.33 mbar cell a- Steady-state polarization and b- Polarization build-up time constant as a function of the discharge intensity. Circles and stars: broadband (2 W) and single mode (0.5 W) pump lasers, both at full power.

weak and one strong discharge at full laser power are presented in Fig.6. The obtained polarizations at high pressures are to our knowledge record values using MEOP. The polarization build-up time weakly depends on  $^3\text{He}$  pressure, in contrast with low-field situations [6, 9].

The specific mechanisms of optical pumping at 1.5 T can be used to account for the observed dynamics. The metastability exchange rate  $\gamma_e$  for a  $2^3\text{S}$  atom is of the order of  $3.75 \times 10^6 \text{ s}^{-1}/\text{mbar}$  [8] while the optical pump-

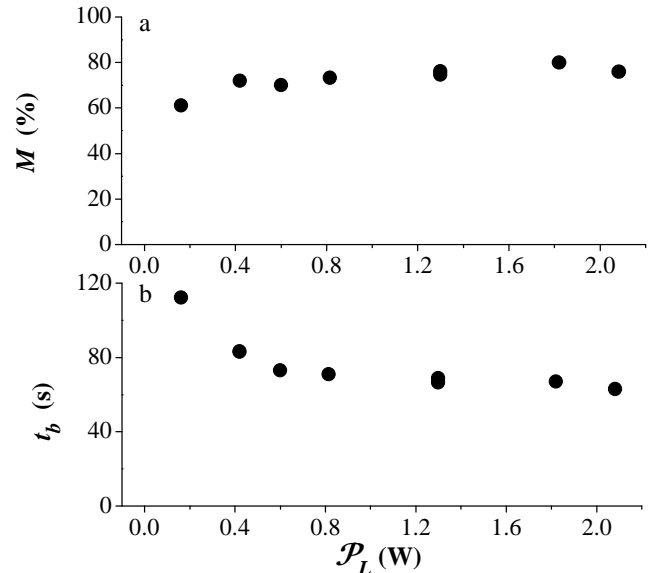


FIG. 5: a- Steady-state polarization and b- Polarization build-up time constant as a function of incident laser power. Data for the broadband (2 W) pump laser in the 1.33 mbar cell for  $T_1=300$  s.

ing rate  $\gamma_p$  is limited by laser power and ultimately by optical saturation and the  $2^3\text{P}$  state finite decay rate  $\gamma = 10^7 \text{ s}^{-1}$ . In our experimental conditions, metastability exchange is the dominant process as far as time scales are concerned. However due to electronic spin conservation, metastability exchange collisions essentially couple sublevels within each pair  $S_L$ ,  $S_M$  or  $S_H$  and a relatively weak optical pumping beam (e.g.  $\gamma_p \geq \epsilon\gamma_e$ ) can efficiently transfer population from the pairs  $S_L$  and  $S_M$  to the pair  $S_H$ . Assuming full pumping of atoms into the pair  $S_H$ , each metastability exchange collision transfers either one unit of angular momentum to the ground state from the  $m_F=3/2$  state or  $(\epsilon-1)$  unit of angular momentum from the  $m_F=1/2$  state. One infers  $t_b = 2T_e/\epsilon$  where  $1/T_e$  is the metastability exchange collision rate for a  $^3\text{He}$  atom in the ground state, proportional to the metastable atoms density. At 300 K,  $1/T_e=1.53 \times 10^{-10} \text{ cm}^3/\text{s} \times n_m$ , yielding  $t_b$  in the range 13-130 s. This prediction is in good agreement with our experimental results and indicates (i) that the dynamics of optical pumping at 1.5 T is indeed intrinsically limited by hyperfine decoupling and (ii) that the build-up times should not vary with gas pressure. At low pressure, our production rates of polarized atoms per unit volume  $R_p=P \times M/t_b$  are lower with respect to standard low-field optical pumping. Nevertheless we can take advantage of the weak pressure dependence of  $M$  and  $t_b$  at 1.5 T to efficiently perform MEOP at higher pressure. In Table I, the obtained rates  $R_p$  for data from Fig.6 are listed with published results for standard low-field conditions and in similar sealed cells. By raising the pressure from 1.33 to 32 mbar, a

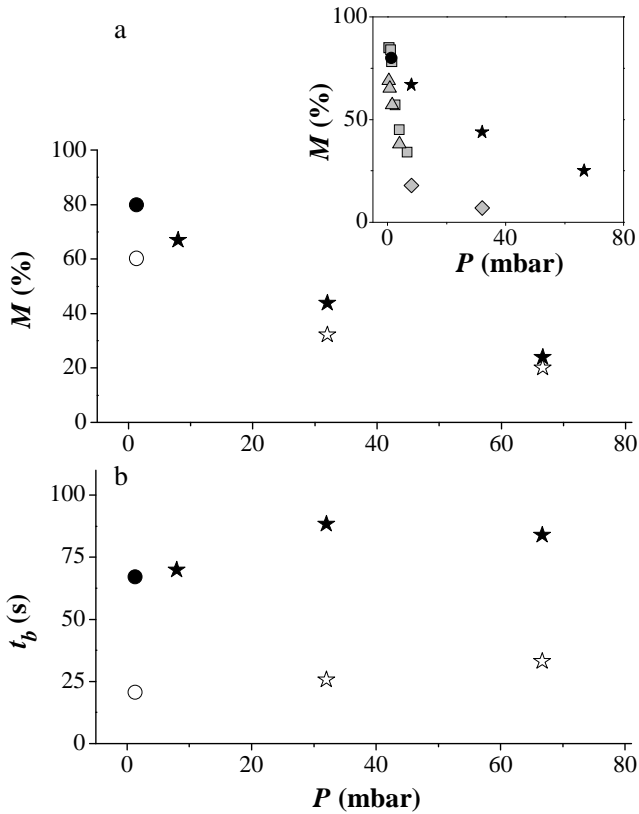


FIG. 6: a- Steady-state polarization, b- Polarization build-up time constant as a function of  $^3\text{He}$  pressure. Circles and stars: broadband (2 W) and single mode (0.5 W) pump lasers, both at full power. Empty symbols are for strong discharges ( $T_1=60, 325$  and  $700$  s for 1.33, 32 and 67 mbar respectively). Full symbols are for weak discharges ( $T_1=300, 2600, 1600$  and  $1300$  s for 1.33, 8, 32 and 67 mbar respectively). *Insert*: High-field data (stars) versus low-field data (all for weak discharges): [6] squares, [5] triangles, and [7] diamonds.

factor 10 is gained for  $R_p$  and fair production rates are recovered. For instance, gas in a 250 cc cell at 32 mbar can be polarized at 40% in 3 minutes. After compression to atmospheric pressure, it is a right amount of gas for imaging the lungs of a guinea pig. For human lung MRI gas accumulation is necessary. Optical pumping at 32 or 67 mbar would considerably simplify the compression stage by reducing the compression ratio from 1:1000 to 1:31 or 1:15. An important point holding at any pressure is that with the large Zeeman splitting in the  $2^3\text{S}$ - $2^3\text{P}$  transition, the magnetic sublevels involved in optical pumping are selected by light frequency and not only by polarization. The high-field optical pumping scheme is therefore extremely robust against polarization impurity of the pumping light. This is a crucial issue for massive production of polarized  $^3\text{He}$  using high laser power, since unwanted polarization would severely limit achieved polarization at low field [10].

We believe that the improvement of the MEOP efficiency at high pressure is due to the inhibition by hyper-

Ref	$P$ (mbar)	$\mathcal{P}_L$ (W)	M(%)	$t_b$ (s)	$R_p$ (mbar/s)
[9]	1	0.05	50 (40)	40 (9)	0.013 (0.047)
[10]	1.33	1.1	56 (39)	11 (2)	0.066 (0.266)
[6]	1.33	4.5	78 (45)	6.5(0.3)	0.160 (2)
1.5 T	1.33	2	80 (60)	67 (20)	0.016 (0.039)
1.5 T	8	0.5	67	70	0.076
1.5 T	32	0.5	44 (32)	88 (26)	0.159 (0.401)
1.5 T	67	0.5	24 (20)	84 (33)	0.191 (0.405)

TABLE I: Steady-state polarization and production rate of polarized atoms per unit volume for published data and for the data in Fig.6. Results in brackets are for strong discharges.  $T_1$  for the published data are: 270 (40) for [9], 400 (10) for [10] and 900 (15) for [6].

fine decoupling of relaxation channels in atomic and/or molecular states in the plasma as indicated by preliminary results at 0.1 T [7]. Our study constitutes a first ensemble of data to identify the most relevant relaxation processes and to understand the fundamental limitations of MEOP. From a practical point of view and in the perspective of a large scale medical use, the development of a polarizer operating at 1.5 T, a widely used field in MRI, could be an attractive choice.

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