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High nuclear polarization in ^{3}He and ^{3}He–^{4}He gas mixtures by optical pumping with a laser diode

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Abstract. New diode lasers delivering 50 mW output power at 1083 nm are shown to be efficient sources for optical pumping of helium. They can polarize nuclei in a ^{3}He gas up to $M = 50\%$ over the pressure range 0.4–1.6 torr. Larger nuclear polarizations M of ^{3}He nuclei, of order 80%, can be obtained in ^{3}He–^{4}He mixtures when the laser frequency is tuned to a ^{4}He line. A standard optical measurement of nuclear polarization M has been extended to the case of ^{3}He–^{4}He mixtures. The effect of various parameters on the steady-state polarization M and on the pumping time T_p is discussed.

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Laser diodes operating at wavelengths close to 1083 nm have recently been developed by Spectra Diode Laboratories [1]. So far, these laser sources are rated to deliver 50 mW of output power in single-mode operation. Taking advantage of the good monochromaticity provided by their DBR (distributed Bragg reflector) technology, one can use these lasers for experiments involving the $2^3S \rightarrow 2^3P$ line of the helium atom [2]. Optical pumping using a laser diode has been demonstrated in ^{4}He [3], and in ^{3}He at low pumping power [4].

In the present work, we have tested the efficiency of these devices to polarize nuclear spins in a ^{3}He gas by optical pumping of the 2^3S metastable state. Laser diodes offer many advantages over the more powerful lamp-pumped LNA lasers (several watts of output power) which are currently used for ^{3}He optical pumping [5]. They are compact, easy to use, commercially available and comparatively inexpensive. Despite their limited power, they are expected to provide fairly high nuclear polarizations: optical pumping through the 2^3S metastable state is a highly nonlinear process, and pumping powers in the milliwatt range generally provide significant polarizations [6–8].

The present work also serves to demonstrate the predicted efficiency of optical pumping in ^{3}He–^{4}He mixtures [8, 9]. The addition of ^{4}He to ^{3}He gas can lead to higher nuclear polarizations of ^{3}He, provided the optical pumping operates on ^{4}He atoms. In this case, the electronic polarization created by the optical pumping in the metastable state of ^{4}He atoms is first transferred to the metastable state of ^{3}He atoms by metastability exchange collisions with ground state ^{3}He atoms. The polarization is then fed into the ground state of ^{3}He atoms via further exchange collisions [8, 9]. The larger light absorption probability of ^{4}He atoms contributes to make this indirect process more efficient in many situations.

We report here results for the steady-state nuclear polarization M_∞, and for the pumping time constant T_p, obtained both for pure ^{3}He and for isotopic mixtures when experimental parameters are varied (such as helium pressure and isotopic composition, discharge intensity, laser characteristics).

1 Optical pumping setup

The experiment is sketched in Fig. 1. The optically pumped helium is enclosed in Pyrex glass cylinders, 5 cm in diameter and 5 cm in length. These cells are filled either with pure ^{3}He gas, or with ^{3}He–^{4}He mixtures at pressures in the torr range. A weak RF discharge is used to populate the 2^3S metastable state. The light emitted by the laser diode is collected by a collimating lens L' ($f = 8$ mm), positioned so as to produce a slightly diverging beam. The pumping beam thus illuminates a large fraction of the cell volume. Since correcting for the elliptical radiation pattern of the laser was found to have almost no effect on pumping characteristics, most experiments are performed with a non-circular beam, having a size in the cell of order 1×3 cm^2. The linear polarization of the light emitted by the diode is transformed into a circular polarization by the

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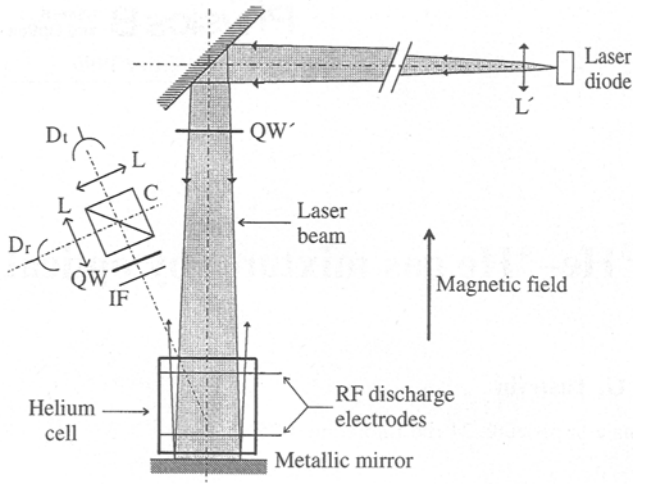

Fig. 1. Sketch of the experimental setup. The helium gas is enclosed in a cylindrical glass cell. A weak RF discharge is sustained by a high-voltage applied to external electrodes. The 1083 nm light emitted by the laser diode is collected by the collimator L' and circularly polarized by the quarter wave plate QW'. The circular polarization of the visible light emitted by the discharge is analyzed by a static polarimeter. It is observed at a small angle with respect to the vertical common axis of the magnetic field and pumping beam; the efect of this angle on the observed polarization is negligible

low-order quartz quarter wave plate QW' and propagates in the cell along a vertical static magnetic field of order 0.3 mT. Since the beam absorption is low in many experimental situations, a metallic mirror is used to reflect the transmitted light back through the cell.

The laser diode has a single-mode operation, with a measured line width of order 3 MHz. The laser frequency can be continuously tuned without mode hopping over the entire fine and hyperfine structure of $2^3S \rightarrow 2^3P$ lines of ^{3}He and ^{4}He atoms. Tuning is obtained by controlling the diode temperature. In addition, a small amplitude high-frequency swing of the laser wavelength can be driven by modulating the intensity of the current fed to the laser diode. This is used to optically pump successively in a rapid sequence various velocity classes of atoms within the Doppler profile.

2 Optical detection of the nuclear polarization

The nuclear polarization M of ^{3}He atoms is measured optically, by monitoring the circular polarization $\mathscr{P}$ of the light emitted by the discharge [10]. The polarimeter which we have used is sketched in Fig. 1 and described in more detail elsewhere [11]. It consists of a quarter wave plate QW and a polarizing beam splitting cube C followed by two photodiodes D_r and D_t which detect the σ_+ and σ_- polarization components of the line selected by the interference filter IF. The discharge amplitude is modulated at 1 kHz with a modulation depth of order 30%, and the modulated components Σ and Δ of the sum and difference of the photocurrents are measured using lock-in amplifiers. The degree of circular polarization of the analyzed light is $\mathscr{P} = \varepsilon\Delta/\Sigma$, where ε would simply be 1 for an ideal polarimeter, but slightly differs from 1 due to various imperfections of the actual device. The present polarimeter offers several advantages over the widely used original device of [10]. It is more compact and easier to make, is not affected by magnetic fields, and is insensitive to ambient or pumping stray light.

Polarization analysis was performed on the red line (at a wavelength of 668 nm), and nuclear polarization M was deduced using the pressure-dependent $M/\mathscr{P}$ ratios measured in [12]. For pure ^{3}He, this method provides a good accuracy of the measured polarizations (better than 1%), except for the weakest discharges at high pressures which emit very little light [11].

In the case of isotopic mixtures, no calibration of optical signals existed so far. Since a fraction of the collected light is emitted by ^{4}He atoms, and hence is unpolarized, a smaller overall circular polarization is expected to be monitored for the same nuclear polarization of ^{3}He atoms. We have performed a series of pulsed NMR measurements in cells containing either pure ^{3}He, or ^{3}He–^{4}He mixtures in order to compare optical signals with an independent determination of the nuclear polarization. The discharge light polarization $\mathscr{P}$ was monitored during the polarization build-up. When a steady state was reached, a $\pi/2$ pulse was applied to the gas in the cell. Free induction signals were then recorded, having a decay time of many seconds [13]. All cells were of identical shape and volume, and the same set of crossed NMR coils was used for all measurements: the initial amplitude of all free induction signals was thus assumed to be simply proportional to MP_3, where P_3 is the ^{3}He partial pressure in the cell. The ratio $[M/\mathscr{P}]_{\text{mix}}$ observed in a ^{3}He–^{4}He mixture of total pressure P_{tot} and ^{3}He concentration $x = P_3/P_{\text{tot}}$ was found to be given by

$$[M/\mathscr{P}]_{\text{mix}}(P_{\text{tot}}, x) = (1/x) \cdot [M/\mathscr{P}]_{\text{pure}}(P_{\text{tot}}),$$

where the ratio $[M/\mathscr{P}]_{\text{pure}}(P_{\text{tot}})$ has the value which would be observed in a pure ^{3}He gas of pressure P_{tot} [12]. Measurements have been performed in a gas of concentration $x = 0.25$, with an accuracy of 5–10%, depending on the pressure. We have also tried a lower concentration ($x = 0.1$), but the accuracy was poorer. To some extent, the optical detection method can thus also be used to measure the nuclear polarization M in an isotopic mixture. Its main drawback is that signal-to-noise ratio deteriorates for low ^{3}He concentrations, but it is very convenient to monitor the time evolution of M, and measure the pumping and relaxation time constants T_p and T_1.

3 Optical pumping results

We present results obtained with five cells listed in Table 1, which differ only by the gas composition and pressure. We have not made an exhaustive study, but have compared polarization dynamics for a limited number of samples under the same laser diode beam size and power (50 mW). For each cell, we have recorded the discharge light polarization $\mathscr{P}$ while the pumping beam was successively applied and blocked. Polarization build-up followed a simple exponential approach towards its asymptotic

Table 1. Gas composition of the studied samples. All cells have the same shape and volume, of order 100 cm^3. Cells P1, P2 and P3 are filled with pure ^{3}He gas, while cells M1 and M2 contain an isotopic mixture with 25% ^{3}He. Cells M1 and M2 contain the same amount of ^{3}He as cells P1 and P2, respectively; cells P3 and M1 are filled with the same total pressure

Cell	^{3}He pressure (torr)	^{4}He pressure (torr)
P1	0.4	0
P2	0.8	0
P3	1.6	0
M1	0.4	1.2
M2	0.8	2.4

value, in contrast to the more complicated increase obtained using a more powerful laser [8]. Pumping time constants T_p, and relaxation time constants T_1 (for the polarization decay induced by the running discharge when the pumping beam is blocked), were deduced from the time evolution of $\mathscr{P}$. The steady-state value of the nuclear polarization, M_∞, was also derived for each recording. For each cell, different discharge intensities were tried, including weak levels close to extinction threshold. All results are plotted in the figures as a function of P_3/T_1. This is a relevant parameter which increases with the discharge level and is related to the loss rate of nuclear magnetization [11].

We have first studied the consequences of laser monochromaticity on the optical pumping efficiency. Pumping with a high spectral density (and/or an unexpanded laser beam) can reduce M_∞, due the saturation of the transition of atoms in a single velocity class [14]. We have compared results obtained using a free running laser diode, and with an applied 20 MHz modulation of the injected current, which caused a laser frequency swing over about 0.4 GHz. This amplitude, a large fraction of the Doppler width (2 GHz fwhm for ^{3}He), as well as the sweep rate, were found sufficient to compensate for the high spectral purity of the laser diode. Over the studied range of discharge intensities, we obtained a relative increase in M_∞ of order 8% in cell P1 and 3% in cell P3 when the laser current was modulated. This pressure dependence is believed to result from the collision-induced orientation transfer between atoms of different velocities: at higher pressures, collisions are efficient enough to fight saturation effects, and no improvement comes from modulating the laser frequency (at least for the moderate power provided by the present diode). In contrast, no significant increase was observed for the isotopic mixtures (cells M1 and M2). For all the results presented below, the laser frequency was modulated only for experiments on pure ^{3}He, but not on isotopic mixtures. Note that the required modulation to obtain the largest polarizations should be determined again if different experimental conditions were used (e.g. more powerful laser, different beam size or cell geometry).

Different pumping transitions were studied for each cell. For pure ^{3}He cells, the laser was tuned on the C_8 or C_9 line ($2^3S_1 \rightarrow 2^3P_0$ transitions of ^{3}He) [7]. For ^{3}He–^{4}He mixtures, pumping on the D_0 line (same $2^3S_1 \rightarrow 2^3P_0$ transition for ^{4}He) was found to be more efficient. Figure 2 shows the nuclear polarization obtained in pure

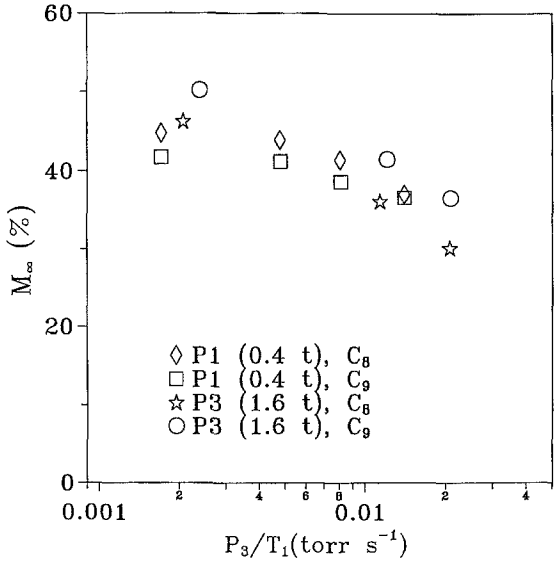

Fig. 2. Steady-state nuclear polarization M_∞ obtained in cells P1 and P3, which contain pure ^{3}He, plotted as a function of the discharge intensity characteristic parameter P_3/T_1. Using the C_8 line is slightly more efficient at low pressure, while the C_9 line should be preferred at higher pressures. The laser power entering the cell is 50 mW

^{3}He with cells P1 and P3. The largest values of M_∞, ranging between 40% and 50%, were obtained for the weakest discharge levels. At the low pressure of 0.4 torr (cell P1), pumping with the C_8 line gives the highest values for M_∞, while the C_9 line is more efficient at higher pressure (cell P3). Similar results had previously been observed and explained [7]. Another interesting feature concerns the pumping times T_p, which have been found to be significantly shorter in all situations when the C_9 line was used, even at low pressure where it resulted in lower values of M_∞. In all of the following, we shall only present results obtained using the C_9 line for pure ^{3}He (cells P1 to P3). For the rather dilute mixtures which we have studied (cells M1 and M2), pumping using the D_0 line was faster and also led to higher polarizations.

Figure 3 displays the steady state polarizations M_∞ obtained in all five cells for various discharge levels. The most striking result is that the addition of ^{4}He (in cells M1 and M2) allows one to obtain significantly larger polarizations, up to 80% despite the limited laser power. This confirms earlier observations [8] and agrees with the calculations of [9].

The pumping time T_p which characterizes the polarization increase towards M_∞ often plays an important role in experiments. It depends both on the discharge intensity, which is characterized by P_3/T_1, and on the laser power, which was kept constant for all experiments. In Fig. 4, we have chosen to plot the combined quantity $M_\infty P_3/T_p$, which is closely related to the rate of production of magnetization. The largest rate was obtained in the mixture cell M1 (note the logarithmic vertical scale). It results from the largest values of M_∞, but also from the fastest pumping times of all five cells. For all cells, larger production rates were obtained when the discharge level was increased. Despite the loss in polarization M_∞ (see

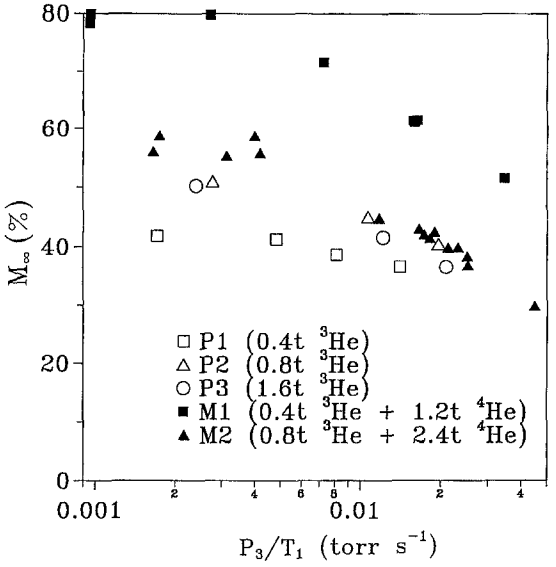

Fig. 3. Comparison of the steady-state nuclear polarizations M_∞ obtained in different cells, plotted as a function of the discharge intensity characteristic parameter P_3/T_1. A higher polarization is obtained in ^{3}He–^{4}He mixtures (closed symbols) than in pure ^{3}He (open symbols). The D_0 line was used for optical pumping of mixtures, the C_9 line for pure ^{3}He

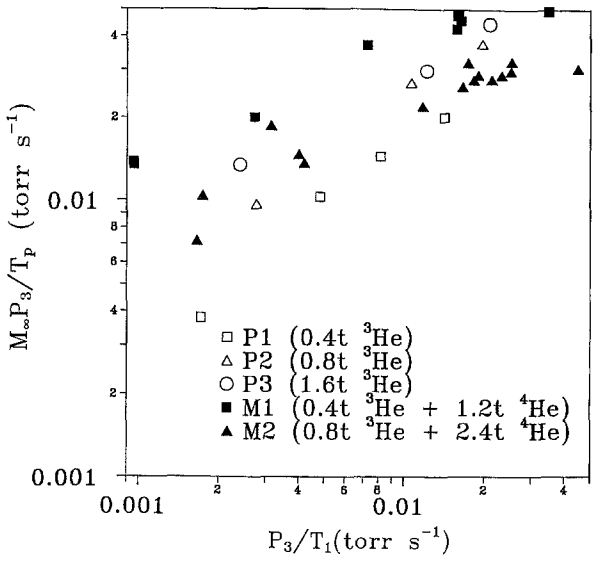

Fig. 4. The measured pumping time T_p and steady-state polarization M_∞ are combined in a single quantity $M_\infty P_3/T_p$ related to the production rate of magnetization, as a function of the discharge intensity characteristic parameter P_3/T_1. For a given cell, $M_\infty P_3/T_p$ increases with discharge intensity, but not as steeply as with a linear dependence (note the two logarithmic scales). Here M_∞ is expressed as a number smaller than 1 instead of percents

Fig. 3), the increase of the metastable state density, and hence of the laser absorption, efficiently reduces the pumping time. Note however that the production rates do not increase so steeply for the strongest discharge levels. The production rate of nuclear magnetization by the optical pumping process obviously depends on the experimental conditions, but also on the polarization M which is desired. For a given value of M/M_∞, the production rate is simply proportional to $M_\infty P_3/T_p$. If, for instance, one is satisfied with only two-thirds of the steady-state polarization M_∞ which is plotted in Fig. 3 (this can be still as high as 50%), an interaction time of order $1.1T_p$ with the pumping beam is sufficient. The graph in Fig. 4 also gives the production rate of magnetization, provided the vertical axis is suitably scaled. For instance, if one chooses again $M/M_\infty = \frac{2}{3}$, the vertical full scale corresponds to a rate of order 10^{17} nuclear spins per second in the 100 cm^3 cells we have studied.

4 Discussion

In summary, we have shown that a 50 mW laser diode operating at 1083 nm is a rather efficient tool to polarize ^{3}He nuclei by direct optical pumping. The use of ^{3}He–^{4}He mixtures can often be preferred if isotopic purity is not required. In mixtures, larger polarizations (up to $M_\infty = 80\%$) can be obtained, with shorter pumping times.

Commercially available laser diodes already have interesting potential applications in the field of polarized ^{3}He. They are clearly the ideal pumping sources for ^{3}He magnetometers [15]. They can also be considered as an easy tool for the production of compressed polarized helium targets [16], with applications to nuclear [17] and high-energy physics, as well as to neutron spin polarimetry [18]. In the latter case, the admixture of ^{4}He to ^{3}He does not cause any problem, ^{4}He atoms being transparent to neutrons. As the factor of merit of the polarized target is the relevant parameter, the potential advantage of using isotopic mixtures (in which significantly larger polarizations are easily obtained) for neutron spin filters is here demonstrated.

All these target experiments are very demanding in terms of nuclear polarizations and pumping time. More powerful laser diodes are highly desirable and, should they become available, would probably replace currently used lamp-pumped LNA lasers. Possible techniques to obtain a more powerful version of the existing laser diode could be to manufacture monolithic master oscillators and power amplifiers, or laser diode arrays. Our results suggest that the limitations introduced in the optical pumping efficiency by the single-mode operation of laser diodes can easily be overridden by a fast modulation of the laser frequency, and we believe that high-power diodes would be very appealing for the future applications.

Let us finally mention the recent application of polarized ^{3}He to magnetic resonance imaging (MRI). Highly polarized noble gases (^{129}Xe or ^{3}He), when introduced into the lungs of an animal [19–21], or inhaled by a living volunteer [22, 23], can provide clear images of the lungs and show brighter images than conventional proton MRI techniques. So far, the polarized ^{3}He gas used in these preliminary experiments was produced either by direct optical pumping of pure ^{3}He followed by a mechanical compression [16, 23], or by spin exchange with optically pumped rubidium [22]. A high-power laser diode array has been used for the optical pumping of rubidium, but the limited efficiency of the spin exchange mechanism has restricted the production rate to about 10^{17} nuclear spins

per second [22]. In the present work, we have obtained comparable production rates with a 50 mW laser diode. However, as in the work of reference [23], a subsequent compression is required for use in MRI. Admixture of ^{4}He would not be a problem for biological imaging applications, and it can clearly improve the efficiency of optical pumping. Since even moderate polarizations can be sufficient if the production rate is fast enough, laser diodes operating at 1083 nm may become a tool very well-suited to such applications in a medical environment.

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