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OPTICAL PUMPING OF HELIUM-3 AT HIGH PRESSURE AND MAGNETIC FIELD*

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At low magnetic field, the efficiency of metastability-exchange optical pumping of helium-3 is known to be optimal for pressures around 1 mbar. We demonstrate on several examples (up to 32 mbar) that operating in a higher magnetic field (here 0.12 T) can significantly increase the nuclear polarisations achieved at higher pressures. Since polarisation measurements cannot be made with the standard technique, we use a general optical method based on absorption measurements at 1083 nm to measure the polarisation of the atoms in the ground state.

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1. Introduction

Highly polarised ^{3}He gas is used in various domains, for instance to prepare polarised targets for nuclear physics experiments [1], to obtain spin filters for cold neutrons [2, 3], or to perform Magnetic Resonance Imaging (MRI) of air spaces in human lungs [4, 5]. All these applications require a very high nuclear polarisation, also called hyper-polarisation since it is orders of magnitude above the Boltzmann equilibrium value (of order 10^{-5}/Tesla at room temperature).

A very efficient and widely used polarisation method relies on optical pumping of the 2^3S metastable state of helium with 1083 nm resonant light [6, 7]. Transfer of nuclear polarisation to atoms in the ground state is

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ensured by metastability exchange collisions. Optical Pumping (OP) is usually performed with a low applied magnetic field (up to a few mT). This field is required only to prevent fast magnetic relaxation of the optically prepared orientation and has negligible effect on the structure of the atomic states. In particular, all Zeeman splittings are much smaller than the Doppler width of the atomic transitions and the pumping light must be circularly polarised to selectively depopulate sublevels and deposit angular momentum in the gas.

OP can provide a high nuclear polarisation, above 80% for optimal conditions [8], but operates efficiently only at low pressure (of order 1 mbar) [9]. Production of dense polarised gas is a key issue for some applications. Polarisation-preserving mechanical compression of the helium gas after OP at low pressure is performed by several research groups using different methods [10–12], but it is a demanding technique and no commercial apparatus can currently be used. Improving the efficiency of OP at higher pressures could facilitate this compression by significantly reducing the requirements on compression ratio and pumping speed. It is also a way to obtain directly larger magnetisation densities. This has been used to perform lung MRI in humans, simply adding a neutral buffer gas to the polarised helium to reach atmospheric pressure and allow inhalation [13].

The efficiency of OP can be improved at high pressures by operation in a magnetic field higher than is commonly used. High field OP in ^{3}He had been previously reported at 0.1 T [14] and 0.6 T [15], but the worthwhile use of a high field (0.1 T) for OP at high pressures (tens of mbar) had not been highlighted until recently [16,17]. A systematic investigation of various processes relevant for OP in non standard conditions (high field and/or high pressure) has been initiated, and there is experimental evidence of molecular formation (metastable He_2 molecules) and of increased relaxation when an intense OP laser light is used in a high pressure plasma [18, 19]. In this article we first discuss various OP situations, with emphasis on the effects of a high magnetic field on the OP process in helium, then give experimental demonstration of the OP improvement obtained using a 0.12 T field in high pressure situations.

2. Known effects of pressure and field on OP in helium

2.1. Standard OP conditions

In order to populate the 2^3S metastable state and perform OP, a plasma discharge is sustained in the helium gas. This constantly produces highly excited states of helium atoms and ions. One of the radiative decay channels ends at the 2^3S metastable state, which in practice may only decay through a collision process:

(i) diffusion to the cell wall and loss of excitation,

(ii) ionising (Penning) collisions [20]

$$\mathrm{He}^* \quad + \quad \mathrm{He}^* \quad \to \quad \mathrm{He} \quad + \quad \mathrm{He}^+ \quad + \quad e^-, \tag{1}$$

(iii) 3-body collisions with conversion into a metastable helium molecule

$$\mathrm{He}^* \quad + \quad 2\mathrm{He} \quad \to \quad \mathrm{He}_2^* \quad + \quad \mathrm{He}\,, \tag{2}$$

(iv) excitation quenching by gas impurities (non-helium atoms or molecules).

In equations (1) and (2), He* refers to either the 2^3S or 2^3P state[1]. In steady state, the replacement of He* atoms having decayed by processes *(i)* to *(iv)* results in an angular momentum loss (*e.g.* through emission of circularly polarised fluorescence light or through depolarising collisions in the highly excited states [21]). On the one hand, this loss can be characterised by the nuclear magnetisation decay time T_1, which is found to decrease for increasing plasma intensities. On the other hand, the steady state density of the 2^3S metastable state atoms increases with the plasma intensity, and so does the OP light absorption and angular momentum deposition rate in the gas. The most favourable plasma conditions, which lead to the highest steady state polarisations, are usually found for weak discharges (for which process *(ii)* is reduced) in a very pure helium gas (^{3}He or helium isotopic mixture) for which process *(iv)* is negligible.

The rates and relative importance of processes *(i)* and *(iii)* strongly depend on the OP cell dimensions and on the gas pressure. When the transverse cell dimension (which governs the lifetimes of all metastable species due to process *(i)*) is of the order of a few cm, an optimal pressure of order 0.5–1 mbar is experimentally found to be most suitable (see Fig. 1). Indeed, the actual optimal plasma and pressure conditions also depend on OP cell shape and size (*e.g.* due to radiation trapping), and on OP laser power and spectral characteristics [22]. This will not be discussed in the following, where we shall only consider the consequences of operation at pressure or magnetic field higher than usual.

2.2. Pressure dependence of OP

At high gas pressure P (above a few mbar) the proportion of atoms in the metastable 2^3S state is reduced since their number density tends to be limited by the non linear process *(ii)*. In addition, the rate of creation of metastable molecular states by process *(iii)* is enhanced with a P^2

[1] In fact, in the presence of an intense OP light, the excited 2^3P state can be almost as populated as the 2^3S metastable state and may play an important role in these collision processes [18, 19].

dependence (equation (2)) and the diffusion lifetime of these molecules linearly increases with P, which results in a higher density of molecular states. These factors tend to reduce strongly the efficiency of OP at high pressure. Fig. 1 displays data already obtained at moderate (0.3 W) [9] or high (several W) [23] laser power. For pure ^{3}He, the polarisation is halved when the pressure is increased from 0.5 to 4 mbar. For the tested isotopic mixture (25% ^{3}He) the dependence on total pressure is much weaker, and the polarisation actually appears to depend only on the ^{3}He *partial* pressure.

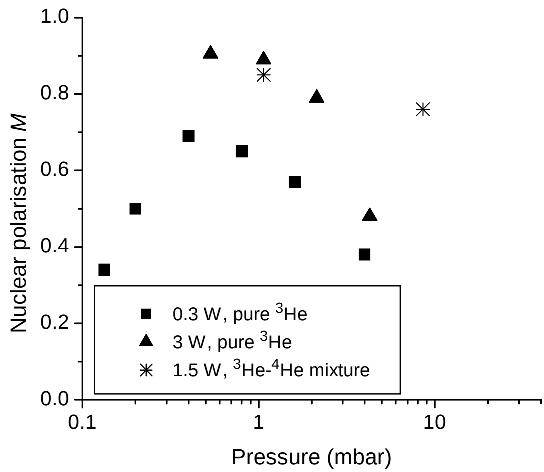

Fig. 1. Steady-state polarisations, obtained by OP at room temperature and low magnetic field, are plotted as a function of the gas pressure. OP was performed at room temperature in 5 cm (diameter) $\times$ 5 cm (length) cylindrical sealed cells filled with either pure ^{3}He, with the OP laser tuned on the C_8 or C_9 transition (the most efficient one is chosen, depending on gas pressure and laser power), or a ^{3}He–^{4}He mixture (25% ^{3}He) with the OP performed on the ^{4}He D_0 line. The low power data (squares) are from reference [9], the other ones from reference [23].

This remains to be fully verified, but it may indicate that only part of the metastable He_2^* molecules (those including a ^{3}He atom) contribute to nuclear relaxation in the plasma.

An even stronger reduction of the efficiency of OP is observed at higher pressures, as will be described and discussed in Section 3.3.

2.3. Field dependence of OP

An important effect of a high enough magnetic field is to strongly reduce the influence of hyperfine coupling in the structures of the different excited levels of helium. In the various atomic and molecular excited states which are populated in the plasma, hyperfine interaction transfers nuclear orientation to electronic spin and orbital orientations. This transfer of orientation has an adverse effect on the OP efficiency by inducing a net loss of nuclear polarisation in the gas. The decoupling effect of an applied field reduces this polarisation loss and may thus significantly improve the OP performance, especially in situations of limited efficiency, such as low temperatures (below) or high pressures (Section 3.3).

At low temperature, a reduced metastability exchange cross section sets a tight bottleneck and strongly limits the efficiency of OP[2] [24–26]. Since the plasma-induced relaxation rate is found to be much faster than the reduced metastability exchange rate, even a strong OP and high polarisation of the 2^3S state result in a limited nuclear polarisation of the ground state: from a few percents at 1 K [27] to 15–20% at 4.2 K [28]. In the latter situation, a field increase up to 40 mT was found to provide a significant improvement in nuclear polarisation, as shown in Fig. 2. Both the relaxation time T_1 and the steady state polarisation increase with the operating field. In this low temperature regime where the orientations in the ground state and the metastable state are only weakly coupled, the observed polarisation increase

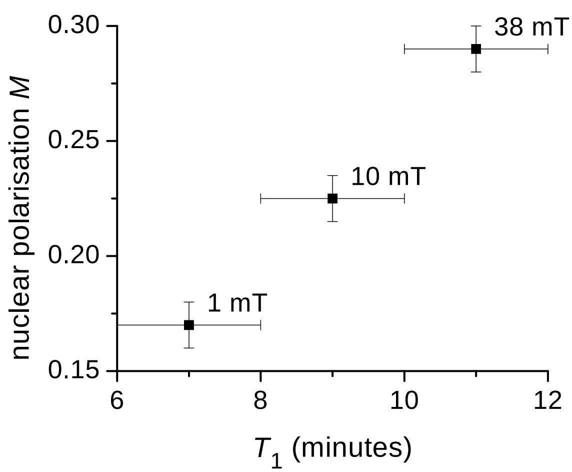

Fig. 2. Steady-state polarisations obtained by OP at 4.2 K are plotted as a function of the measured relaxation time T_1 for three values of the field B. OP is performed in a 5 cm (diameter) × 3.5 cm (length) sealed cell, filled at room temperature with 1.33 mbar of ^{3}He and 6 mbar of H_2 (to form a solid H_2 coating and prevent wall relaxation). The OP laser (100 mW) is tuned on the C_5 transition, the most efficient one in these conditions (data from [28]).

[2] This was extensively studied [26,28] in an attempt to directly obtain high polarisations in a quantum fluid (a helium vapour or liquid, at low enough temperature for quantum statistics to play an essential part in thermodynamic and transport properties of the fluid).

(proportional to T_1) can be directly attributed to the reduced relaxation assuming that OP and exchange processes are not significantly affected by the field increase (a reasonable assumption for these moderate field values [17]).

The OP conditions at high pressure are actually quite different since very frequent metastability exchange collisions strongly couple the orientation in the 2^3S state to that of the ground state. Still, it is not surprising that a significant improvement is obtained by suppressing relaxation channels in high field [16], even if the details of the involved relaxation processes remain to be fully elucidated.

3. New OP results at high pressure and high field

As a demonstration of the improvement of OP obtained at high pressure when operating in a high magnetic field, we report measurements performed in similar conditions at 1 mT and 0.115 T, both in pure ^{3}He and in an isotopic mixture.

3.1. Experimental setup

The experiment arrangement is sketched in Fig. 3. The helium is enclosed in sealed cylindrical Pyrex glass cells, 5 cm in diameter and 5 cm in length. Results presented here have been recorded in 3 cells filled with 8 mbar or 32 mbar of pure ^{3}He, or 32 mbar of helium mixture (25% ^{3}He, 75% ^{4}He). A weak RF discharge ($<$ 1 W at 3 MHz) sustained by means of external electrodes is used to populate the 2^3S state in the cell. The magnetic field B is produced by an air core resistive magnet of sufficient homogeneity over the total cell volume to induce negligible magnetic relaxation in these OP experiments [17].

The probe laser source is a 50 mW laser diode (6702-H1, formerly manufactured by Spectra Diode Laboratories). Its output is collimated into a quasi-parallel beam using an anti-reflection coated lens (f = 8 mm), and attenuated to provide a weak probe beam. It passes across the cell perpendicular to B with linear polarisation such that the σ and π polarisation components are equal. The absorption of the probe beam components is measured using a modulation technique. The discharge intensity is modulated at a low enough frequency (~100 Hz) for the density of the absorbing atoms 2^3S to follow the modulation, and the σ and π intensities are analysed using lock-in amplifiers. The average values of the transmitted probe intensities are also recorded, and used to normalise the absorption measurements. This eliminates errors due to laser intensity changes and strongly reduces the effects of optical thickness of the gas [18].

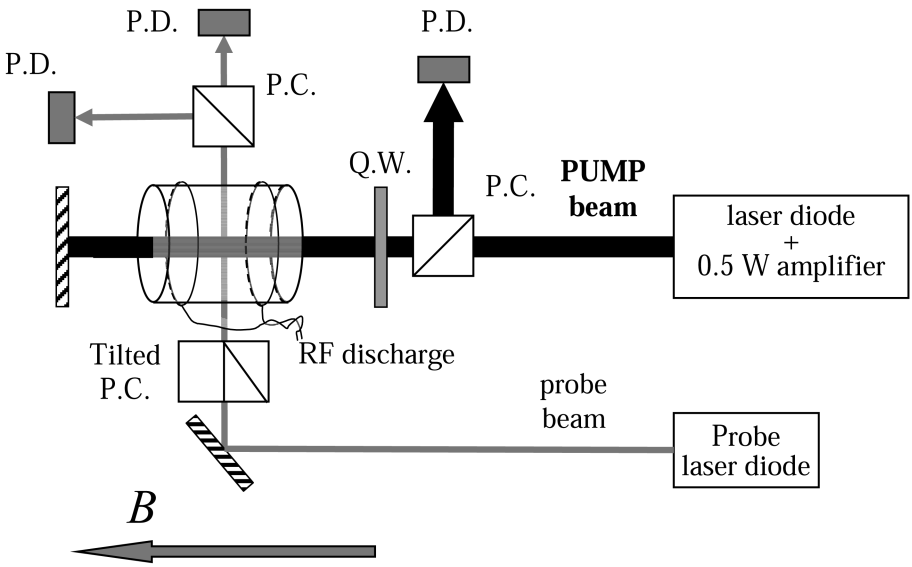

Fig. 3. The main elements of the OP experiment are shown (not to scale). The pump beam, parallel to the magnetic field B, is circularly polarised using a linear Polarising Cube (P.C.) and a Quarter-Wave plate (Q.W.). The absorption of the pump beam is monitored by a Photo-Diode (P.D.) after a double pass through the cell. The transverse probe beam is prepared with σ and π polarisation components which are separated after crossing the cell and simultaneously recorded.

The OP laser used for these experiments is a second laser diode amplified using a 0.5 W fibre amplifier [33] (YAM-1083-500, manufactured by IPG Photonics). The pump beam is expanded (diameter ~ 3 cm) to match the plasma distribution in the cells, and back-reflected after a first pass in the cell to take advantage of the usually weak light absorption.

3.2. Optical detection method

In the standard optical detection technique [8, 29, 30], the circular polarisation of a chosen helium spectral line emitted by the plasma is measured and the nuclear polarisation M of the *ground state* of ^{3}He is inferred. This technique relies on hyperfine coupling to transfer angular momentum from nuclear to electronic spins in the excited state which emits the monitored spectral line. The decoupling effect of an applied magnetic field unfortunately reduces the efficiency of this angular momentum transfer, which is also sensitive to depolarising collisions. This technique must then be used at low fields ($\lesssim 10$ mT), low gas pressures ($\lesssim 5$ mbar) and limited ^{4}He concentrations ($\lesssim 50\,\%$ ^{4}He) to avoid a significant sensitivity loss ($\div 2$ for each of the quoted limits).

Other optical methods, which rely on absorption measurements on the 2^3S–2^3P transition, have been successfully used to quantitatively determine the nuclear polarisation of ^{3}He [6, 8, 31, 32]. They provide information both on the total number density of atoms in the 2^3S state and on the relative populations of the probed sublevels. In usual situations[3], the population distribution in the 2^3S state is strongly coupled by metastability exchange collisions to that in the ground state. These populations would exactly be ruled by a spin temperature distribution in the absence of OP or relaxation processes, both in a low [7] and a high [17] magnetic field.

When two absorption measurements directly probe two populations of atoms in the 2^3S state, the derivation of M is a straightforward procedure. This is for instance the case at low field when the line C_8 or D_0 is probed with σ_+ and σ_- circular polarisations, or at high enough magnetic fields for the Zeeman shifts to remove all level degeneracies ($B \gtrsim 50\,\mathrm{mT}$ [17]). When transitions simultaneously probe several sublevels (*e.g.* in low field with σ polarisation on any line, with any polarisation on line C_9, *etc* ...), the measurements of two independent combinations of populations can still be used to infer the nuclear polarisation M, but specific calculations are then required [8]. Similar results are indeed obtained in an isotopic mixture when ^{4}He atoms are probed to measure relative populations among the three sublevels in the 2^3S state [17].

In this experiment, absorption spectra of the probe beam are recorded for σ and π polarisations over the C_8–C_9 transitions (for pure ^{3}He) or the D_0 transition (for mixtures), both for $M = 0$ and in steady state OP situations. Recording both polarisation channels is required to infer the value of M only in low field situations. The high field spectra provide a redundant determination of population ratios, which is used to check for the consistency of the measurements.

3.3. Experimental results

Fig. 4 displays an example of absorption spectra obtained at 0.115 T in the 32 mbar cell filled with pure ^{3}He gas, for the π-polarised probe beam component. As the laser frequency is tuned over the 2^3S–2^3P$_0$ transition, four resonance lines (two for C_8 and two for C_9) are recorded, which correspond to optical transitions between hyperfine sublevels of identical angular momentum projections m_F along the field axis. All the peak amplitudes are precisely measured. The probe beam intensity is here too weak to optically pump the metastable state. The data redundancy can be used to check

[3] This would not hold at very low pressures, nor in the low temperature conditions discussed in Section 2.3, but is expected to be very well verified for pressures above a few mbar at room temperature.

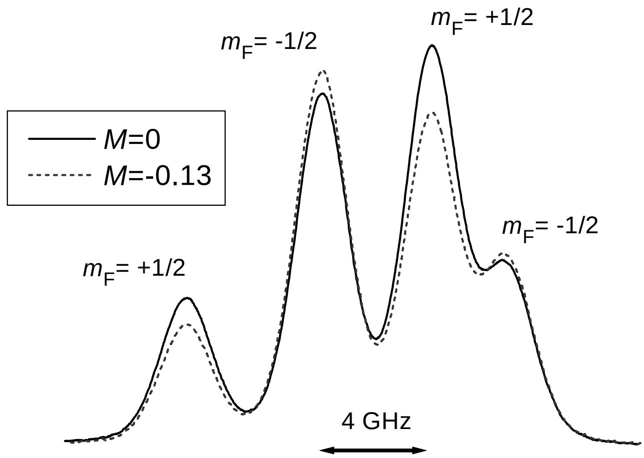

Fig. 4. Absorption spectra of the π component of the transverse probe, tuned to the C_8 and C_9 resonance lines of ^{3}He, in the cell filled with 32 mbar of pure ^{3}He gas. The applied magnetic field is B=0.115 T, and optical pumping is performed with a C_9 pump beam and σ_- circular polarisation at moderate discharge intensity. The nuclear polarisation M is deduced from the measured the peak amplitudes at null polarisation (solid line) and steady-state polarisation (dotted line). The $m_F = +1/2$ states are here depleted, and the resulting ^{3}He nuclear orientation is -0.13 [17].

that the OP beam introduces no spurious population differences between pumped and unpumped metastable sublevels (*i.e.* no local over-polarisation of the 2^3S state as compared to the ground state). This effect of OP on the metastable populations has been discussed both at low and high fields [8,17], and is indeed not expected to be significant in the present magnetic field and pressure ranges. The population ratio of two adjacent hyperfine sublevels is used to infer the spin temperature, and hence the nuclear polarisation M. The total number of 2^3S atoms can also been extracted from these absorption amplitudes. The accuracy of such transverse probe beam absorption measurements is much worse at low field, where the $\pm\, m_F$ sublevels are degenerate in each hyperfine level. In this case, the absorption rates of the σ and π components are even functions of M, and the registered population changes scale like M^2 only. Accurate results can be obtained with a longitudinal probe beam from the comparison of absorption rates for σ_+ and σ_- polarisations.

The results of all polarisation measurements are presented in Table I.

TABLE I

Table 1. Typical steady state nuclear polarisations achieved with the 0.5 W monomode laser for low discharge intensities. OP is performed on the C_9 line with σ_- polarisation in pure ^{3}He, and on the D_0 line with σ_- polarisation in the helium mixture.

	$B = 1\,\mathrm{mT}$	$B = 0.115\,\mathrm{T}$
8 mbar ^{3}He	18%	28%
32 mbar ^{3}He	7%	14%
8 mbar ^{3}He + 24 mbar ^{4}He	14%	23%

At low field, the pressure dependence of the polarisation in our data is consistent with the previously reported one (see Fig. 5, insert). A significant increase of the steady state nuclear polarisations achieved by OP is demonstrated at 0.115 T for all cells. In comparison with results obtained at 1 mT, M is found to be 1.6 times higher at 8 mbar in pure ^{3}He or at 32 mbar in

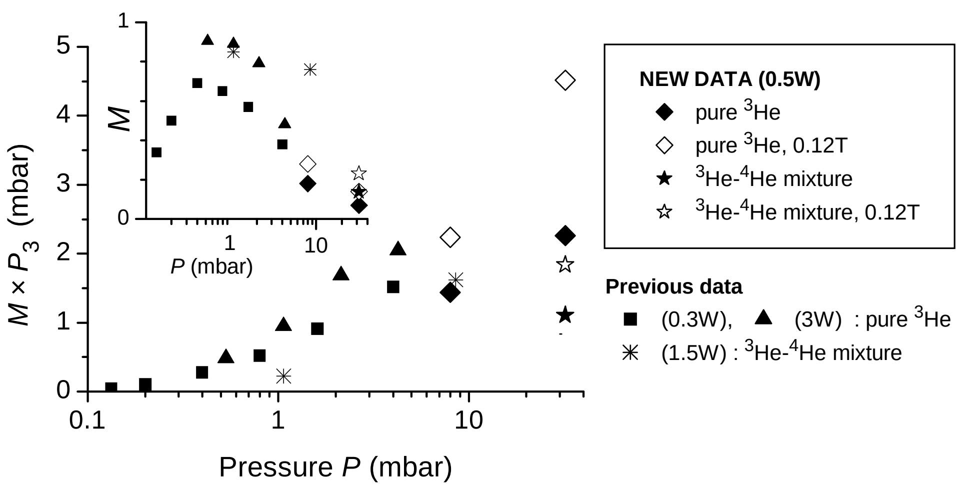

Fig. 5. Insert: The nuclear polarisation measured at 1 mT (filled diamonds for pure ^{3}He, filled star for the isotopic mixture) and 0.115 T (open diamonds and open star, respectively) for the three cells are compared to the previous data (from Fig. 1). Main plot: The total nuclear magnetisation is proportional both to the polarisation M and to the ^{3}He pressure (P_3) in the cell. Improvement is particularly important at 0.115 T in pure ^{3}He.

the isotopic mixture with 8 mbar ^{3}He partial pressure, and 2 times higher at 32 mbar in pure ^{3}He. The improved OP efficiency at high field is further emphasized in Fig. 5 (main plot). The ^{3}He nuclear magnetisation actually produced in the cell, which combines the variation of the polarisation and of the ^{3}He content, steadily increases with pressure. This net gain would directly result in a comparable enhancement of the NMR signal, hence of the image quality, for applications in lung MRI for instance [34].

4. Conclusion

These positive results demonstrate the potential benefit of high field operation for metastability exchange OP at pressures higher than a few mbar. Hyperfine decoupling in the 2^3S state [17] is expected to set an ultimate limit to the intensity of the applied magnetic field, beyond which optical pumping would mainly create electronic polarisation. A systematic study is thus needed to determine the optimal operating field for maximum efficiency as a function of the experimental conditions and requirements. On-going work aims at a detailed analysis of steady state polarisations at higher magnetic field as a function of pressure, isotopic ratio, discharge intensity and laser characteristics. By optically monitoring the evolution of metastable populations, the polarisation process can also be dynamically analysed. A study of the kinetics of OP, focussed on the influence of the magnetic field on both the pumping rates and the relaxation rates, is in progress.

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