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## NUCLEAR POLARIZATION IN GASEOUS $^3\text{He}$ BY OPTICAL PUMPING

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Optical pumping can provide significant nuclear polarizations in gaseous  $^3\text{He}$ . This method, developed by Colegrove, Scheerer and Walters 20 years ago, can be applied to the realization of polarized targets for nuclear physics or neutron spin state filters. Moreover, highly polarized discharges in gaseous helium can be used as sources of polarized beams of atomic or molecular ions or electrons.

Some progress has been made recently to obtain higher nuclear polarizations in gaseous  $^3\text{He}$  at a higher density. Laser optical pumping can produce polarizations over 60% or more in a gas at room temperature. This can be combined with a polarization transfer technique, in which a laser is used to pump  $^3\text{He}$  in a cell at room temperature, while another cell at a few degrees Kelvin is polarized by diffusion of the atoms through a long connecting tube. Cryogenic coatings, made of solid hydrogen frozen on the coldest parts of the inside wall of the container, reduce very effectively the fast nuclear relaxation which would occur on bare pyrex walls at low temperatures. Nuclear polarizations of the order of 50% have been obtained in a relatively dense gas ( $n \approx 10^{18} \text{ cm}^{-3}$ ) at  $T \leq 4 \text{ K}$ . Higher densities should be attainable by optical pumping at higher gas pressures and, possibly, by subsequent compression of the gas at low temperature.

### INTRODUCTION

Several methods can be used to obtain a significant nuclear polarization in gaseous or liquid  $^3\text{He}$ . The simplest idea in principle is probably to use the so-called 'brute force technique', where the fluid is submitted to a high magnetic field at low temperatures. Unfortunately, this method is not very efficient for polarizing the gas because the system solidifies before one reaches temperatures which are low enough to obtain high polarizations. For example, in a field of 10 Tesla ( $10^5$  Gauss) the nuclear polarization is only  $\approx 4\%$  at  $T = 0.2 \text{ K}$ , where the maximum number density in the gas is  $n \approx 10^{15} \text{ atoms/cm}^3$ . The efficiency is even worse if the method is applied to pure liquid  $^3\text{He}$ , which is a degenerate Fermi system with constant and relatively weak magnetic susceptibility below  $T \approx 0.4 \text{ K}$ <sup>1</sup>: For example, in the same experimental conditions as above, the nuclear polarization would be only  $\approx 2\%$  and remain practically constant at lower temperatures. On the other hand, the brute force method can be applied to solid  $^3\text{He}$  or very dilute solutions of  $^3\text{He}$  in liquid  $^4\text{He}$ , which are weakly degenerate systems; large nuclear polarizations can be obtained in a magnetic field of 5-10 tesla and at temperatures of the order of 10 mK. Castaing and Nozières proposed to produce polarized liquid  $^3\text{He}$  by fast melting of a polarized solid<sup>2</sup> and several groups have obtained in this way large nuclear polarizations,<sup>3</sup> as high as 80% in

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some cases.† One can also imagine methods for polarizing liquid  $^3\text{He}$  which rely on surface phenomena, such as adsorption of liquid  $^3\text{He}$  on zeolites<sup>4</sup> (the magnetic susceptibility of a two dimension adsorbed phase of  $^3\text{He}$  may be very different from that of bulk  $^3\text{He}$ <sup>5</sup>) or coupling of the  $^3\text{He}$  spins with  $^{19}\text{F}$  spins in small fluorocarbon particles.<sup>6,7</sup>

The object of this article is to describe an alternative method of polarization, optical pumping; unlike the preceding techniques, it does not necessarily require the use of low temperatures, although it can be combined with them.<sup>8,9</sup> After a general description of the method and of its possibilities, we shall discuss a number of potential applications to the production of polarized beams or targets. We shall then report on some recent progress on the way to higher polarizations and densities, which now make the method more attractive than a few years ago.

A review of polarized  $^3\text{He}$  targets and ion sources by optical pumping can be found in Ref. 10.

## OPTICAL PUMPING IN HELIUM

Optical pumping is a method of transferring angular momentum from the photons of a pump beam to the atoms.<sup>11,12</sup> Usually the pumping transitions correspond to the optical resonances of the atoms, but this is not practical for helium, because its resonance wavelength falls much too far in the ultraviolet. An indirect optical pumping scheme is therefore necessary, such as optical pumping of an alkali vapour and polarization transfer to  $^3\text{He}$  by collisions.<sup>13</sup> At the moment the most efficient scheme seems to be that developed by Colegrove, Scheerer and Walters.<sup>14</sup> Their method involves optical pumping of the  $2^3\text{S}_1$  metastable level of helium at the wavelength  $\lambda = 1.08\mu$  ( $2^3\text{S} \leftrightarrow 2^3\text{P}$  transition), followed by metastability exchange collisions which strongly couple the nuclear polarizations of the metastable and ground states (Figure 1). Some population of the  $2^3\text{S}_1$  level can be produced by a RF discharge in the gas, which is usually maintained near the weakest possible level where the production of metastable atoms is sufficient, but the nuclear relaxation of the ground state by the various products of the discharge (electrons, ions, molecules, etc. . .) is not too rapid.‡

The number densities of the gas where optical pumping is efficient are fixed by the requirement of a reasonably homogeneous weak discharge in the gas and a long lifetime of the metastable levels (if the gas purity is sufficient, the main source of loss for the metastable depopulation is diffusion of the atoms to the walls, where they are deexcited by collision). In practice, this corresponds to number densities ranging from  $3 \times 10^{15}$  to  $3 \times 10^{17}$  atoms/cm<sup>3</sup>, the optimum for high polarization being close to  $10^{16}$  atoms/cm<sup>3</sup>.

This method has now been used for years with strong discharge lamps, often excited by 100 watts or more of RF power, as pumping light sources. Despite all the efforts of various groups to increase the light intensity and the pumping efficiency (spectral purity) of these sources, nuclear polarizations much greater than 20% have not been obtained.<sup>15-17</sup> Another way to increase the nuclear

† This method provides only transient nuclear polarizations, which relax to a much smaller value in a time ranging from  $\approx 10$  s to about 300 s, depending on the experimental conditions.

‡ In practice, the nuclear relaxation time in the active discharge is of the order of a few minutes, instead of several hours (limited by the wall relaxation) in the absence of a discharge.

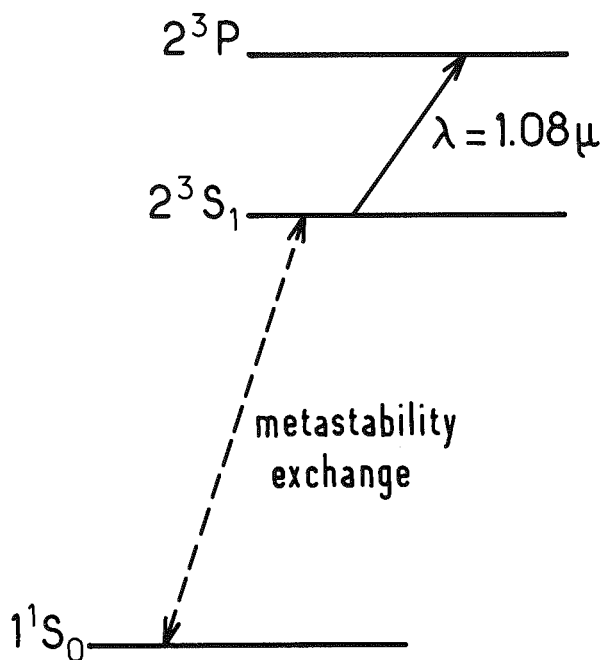


FIGURE 1 Scheme of the helium levels involved in  $^3\text{He}$  optical pumping. The metastable  $2^3S_1$  atoms are oriented by optical pumping with the  $\lambda = 1.08 \mu$  line. A rapid transfer of orientation towards the ground state atoms is ensured by metastability exchange collisions.

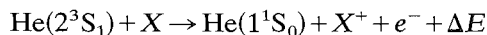
polarizations would be to find a way to produce the metastable atoms without introducing too much nuclear relaxation of the ground state, but no solution to this experimental problem seems to have been found yet.

Optical pumping can be performed not only on a gas, but also on a beam of metastable  $^3\text{He}$  or  $^4\text{He}$  atoms. For  $^3\text{He}$ , this situation is actually simpler than pumping an active discharge or even a flowing afterglow; the reason is the strong reduction of the effects of metastability exchange collisions with ground state atoms, which otherwise introduce strong couplings between the population of the Zeeman and hyperfine levels of the metastable state.<sup>14,18</sup> Higher spin polarizations in the metastable level can therefore be obtained more easily in this case.

Two recent improvements have extended the possibility of the optical pumping method; the design of a relatively powerful laser to generate the desired wavelength  $\lambda = 1.08 \mu$ ,<sup>19</sup> and the use of nuclear polarization transfer techniques between cells at different temperatures.<sup>20</sup> The former allows one to obtain nuclear polarization of the order of 70% by optical pumping. The latter has been used to obtain high nuclear polarizations in a denser gas ( $n \approx 10^{18}$  at/cm<sup>3</sup> in a preliminary experiment) at low temperatures. These experiments will be described in more detail below; their main motivation was the study of the quantum properties of spin-polarized  $^3\text{He}$  (or  $^3\text{He}\uparrow$ ) at low temperatures:<sup>21</sup> enhanced viscosity or heat conduction, increased saturating vapour pressure of the liquid, and spin waves.<sup>22</sup> There are nevertheless several other potential applications of nuclear polarizations created by optical pumping in gaseous helium, as we shall see in the next section.

## POSSIBLE APPLICATIONS

In this section, we briefly review possible uses of highly polarized gaseous helium discharge as source of polarized particles. We include in the discussion both  $^3\text{He}$  and  $^4\text{He}$ .  $^3\text{He}$  can have nuclear polarization in the ground state and both nuclear and electronic spin polarizations in the metastable state (where they are coupled by a strong hyperfine coupling). The only polarization possible for  $^4\text{He}$  is electronic spin polarization in the metastable state. Provided that any of these levels is oriented, the polarization is rapidly transferred by various types of collisions to other species:  $\text{He}^+$  ions,<sup>23,24</sup> electrons,<sup>25-27</sup> molecules and molecular ions.<sup>28,29</sup>† The number and variety of the polarized species can even be increased if some impurities are added in the gas. The high internal energy of the metastable  $2^3\text{S}$  atoms (20 eV) is sufficient to ionize all atoms but He and Ne, so that Penning collisions with some atomic or molecular impurity X:



are energetically possible and often have a large cross section. After the collision, the impurity ion  $\text{X}^+$  can be left either in its electronic ground state, or in some excited state. An interesting feature of these collisions is that they transfer the polarization of the metastable state to both the ion  $\text{X}^+$  and the outgoing electron. The reason for this is the pure spin character of the initial polarization, and the fact that collision processes are generally much too short (e.g.  $\approx 10^{-12}$  s) to allow for any appreciable magnetic interaction of the spins (this spin conservation rule is sometimes referred to as the Wigner spin rule<sup>30</sup>). The injection of the impurities is generally combined with the use of a flowing afterglow technique,<sup>31</sup> so that the optical pumping region is not perturbed by their effect (reduction of the metastable density due to the Penning collisions). In this way, L. D. Schearer and his colleagues were able to polarize ions of metallic atoms,<sup>32</sup> electrons<sup>33</sup> and molecular ions,<sup>34,35</sup> and to do various spectroscopic experiments.<sup>36</sup> Similar experiments were also performed by Hamel *et al.*,<sup>37,38</sup> as well as by Zhitnikov and his group.<sup>39,40</sup>

Probably more interesting from the point of view of nuclear physics techniques is the fact that charged species can be extracted electrostatically from the discharge without losing their spin polarization.<sup>22,41</sup> In this way an intense source of polarized electrons using laser induced optical pumping has recently been built,<sup>42</sup> and the performances obtained compare well with other electron spin polarization techniques such as GaAs sources.

Another application that does not require that the polarized species be extracted from the active discharge is the use of the ground state  $^3\text{He}$  atoms as a polarized target into which a beam of particles ( $\alpha$  particles for instance) can be shot to produce nuclear reactions. The first team to demonstrate this possibility was the group at Rice University<sup>43</sup> with a  $\text{He}^{++}$  beam (energy  $\approx 6$  to 7 MeV). This polarized target, with a nuclear polarization of the order of 10–15%, was later used with a beam of deuterons to measure left-right asymmetries in the production of protons. Several other groups subsequently started to build optically pumped  $^3\text{He}$  targets for nuclear physics, especially a group in Toronto<sup>44</sup> who developed a method to compress the polarized gas with a Toepler pump.<sup>45</sup> In this way, the number density of the gas was increased up to  $\sim 5 \times 10^{18} \text{ cm}^{-3}$ , with a

† Of course, all these transfer processes take part in the depolarization of the He atoms in a discharge.

nuclear polarization  $\approx 3\%$ . Unfortunately, all these experiments were done before the time when a laser at  $\lambda = 1.08\mu$  became available as a source for optical pumping, and this explains why the nuclear polarizations obtained were relatively limited. Very recently, experiments have been reported by a group at the University of Manitoba.<sup>46</sup> Although no laser was used, nuclear polarizations slightly greater than 20% were obtained in the target, but no gas compression was performed.

A polarized target of  $^3\text{He}$  could also be used as a neutron spin state filter to produce polarized neutron beams. It is known that the cross section for the capture of a thermal neutron by a  $^3\text{He}$  atom is strongly spin dependent.<sup>4</sup> This is because the capture involves the formation of an excited state of  $^4\text{He}$  with zero angular momentum. The advantage of an absorption method, as compared to other neutron polarization methods such as reflexion on surfaces of saturated ferromagnetic materials, would be a broader angular and energy acceptance.<sup>47,48</sup> Polarized targets of  $^{151}\text{Eu}$  and  $^{149}\text{Sm}$  have already been used for that purpose at very low temperatures, of the order of 10 mK.<sup>49</sup> It has also been pointed out recently<sup>50</sup> that the strong spin dependence of the absorption of ultra-cold neutrons by  $^3\text{He}$  nuclei could be used in a very precise search for the neutron electric dipole moment.

Another potential application, although possibly rather speculative, has been the object of much interest recently. It has been known for a long time that polarizing the nuclei in fusion plasmas would be advantageous in several cases.<sup>51</sup> For example, polarizing both the helium and deuteron nuclei in a D- $^3\text{He}$  reactor would at the same time enhance the D- $^3\text{He}$  cross section by 50%, and strongly suppress the D-D reaction and its undesirable neutron output. It was nevertheless thought that the nuclear spin relaxation time in a hot plasma would be much too short to conserve any polarization. Only recently, more precise calculation of the relaxation times showed that they should actually be much longer than previously expected,<sup>52,53</sup> so that experiments might be possible. Even if a significant change of the efficiency of a real fusion reactor seems to be a rather remote future prospect, applications such as plasma diagnostics<sup>54</sup> could be a more immediate project. It has recently been suggested<sup>55</sup> that an intense beam of polarized metastable  $^3\text{He}$  atoms be injected into a fusion reactor.†

Let us finally mention that the optical pumping method can be used to polarize other phases than the gaseous phase. This can be done indirectly by creating a nuclear polarization in the gas first, and then polarizing the liquid by atom exchange with the gas. This possibility was demonstrated by McAdams and Walters in 1967,<sup>58</sup> although the polarization obtained was rather low. As we shall discuss below, there is hope to improve these results in the near future.

## LASER OPTICAL PUMPING OF $^3\text{He}$

As mentioned above, the efficiency of the optical pumping method can be significantly improved by the use of an infrared laser as pumping source on the  $\lambda = 1.08\mu$  transition of helium. We shall discuss in this section a model to calculate the nuclear polarization  $M$  in the ground state as a function of the laser intensity  $I$  and its frequency; then, we shall compare the predictions of this model with experimental results.

† Highly polarized beams of metastable helium atoms have already been obtained<sup>56</sup> and used for studies of electron ejection from solid surfaces.<sup>57</sup>

The kinetics and efficiency of optical pumping in  $^3\text{He}$  was already studied in the first articles on this subject.<sup>14</sup> More detailed and quantitative calculations were later performed by Daniels and Timsit.<sup>15</sup> The model we discuss here is a slight generalization of this work: with laser optical pumping it is possible to excite any component of the pumping line and the pumping time can be short compared to the metastability exchange time for the  $2^3\text{S}$  level.

As shown in Figure 2-a, the metastable  $2^3\text{S}_1$  level and the  $2^3\text{P}$  level are split respectively into 2 and 5 fine and hyperfine structure sublevels; the 9 resulting components of the  $\lambda = 1.08\mu$  lines are labelled  $C_1, C_2, \dots, C_9$ , in Figure 2-b.

The total number of Zeeman sublevels inside the metastable state is 6 (2 and 4 for the  $F=1/2$  and  $F=3/2$  sublevels respectively). A simple model can be built by writing the evolution in time of the 6 corresponding populations, coupled by metastability exchange collisions to the populations of the 2 Zeeman sublevels in the ground level  $1^1\text{S}_0$ . The effect of three other physical processes will be included in the model; absorption of photons of the laser beam, which is supposed to be circularly polarized; spontaneous emission which brings back the atoms from the  $2^3\text{P}$  level to the metastable level; depolarizing collisions acting inside the  $1^1\text{S}_0$ ,  $2^3\text{S}_1$  and  $2^3\text{P}$  levels.

As discussed in Reference 59, metastability exchange collisions are short processes during which excitation of the electronic cloud is transferred from one nucleus to the other, but the nuclear spins themselves are unaffected; the nuclear orientation of the metastable atom is totally transferred to the ground state atom, and conversely; the electronic spin orientation of the metastable atom is unchanged by the collision process itself (but can be changed afterwards under the effect of the hyperfine coupling). As shown in Reference 18, the metastability exchange collisions introduce not only a coupling between the populations of the metastable level and those of the ground state, but also between the metastable populations themselves; moreover, this coupling is in general non linear (it can be linearized at low polarizations, but we do not want to limit ourselves to this case). Consequently, the equations of evolution of the model cannot be solved analytically, and a numerical calculation is necessary.

The effect of depolarizing collisions inside the ground state (collisions against various products of the discharge, such as electrons, ions, molecules) can be treated phenomenologically by introducing a relaxation probability ( $1/T_r$ ) of the nuclear polarization  $M$  of the ground state. Atom-wall collisions also contribute to ( $1/T_r$ ) but, at room temperature, this is generally a negligible effect. As for depolarizing collisions inside the  $2^3\text{S}$  and  $2^3\text{P}$  levels, they cannot be treated simply in a similar way by the introduction of relaxation probabilities ( $1/\tau_r$ ) and ( $1/\tau'_r$ ). The reason is that these levels possess a nuclear angular momentum  $\mathbf{I}$ , an electronic spin angular momentum  $\mathbf{S}$ , and an electronic orbital angular momentum  $\mathbf{L}$  (for the  $2^3\text{P}$  level), which are affected very differently by the collisions;  $\langle \mathbf{L} \rangle$  is efficiently depolarized,  $\langle \mathbf{S} \rangle$  at a much lower rate, and  $\langle \mathbf{I} \rangle$  almost totally conserved. To simplify the model, we shall ignore the effect of depolarizing collisions inside the  $2^3\text{S}$  level, which does not possess any  $\mathbf{L}$  angular momentum. Accounting for the effect of depolarizing,  $\mathbf{J}$  and  $\mathbf{F}$  changing collisions inside the  $2^3\text{P}$  level would be a complicated task, even if only the depolarization of  $\mathbf{L}$  were considered, due to the number of sublevels involved (18 including fine, hyperfine and Zeeman structures). We shall thus only study two extreme cases, corresponding respectively to very low and very high pressures; no depolarization at all, or complete thermalization of the populations of the sublevels before spontaneous emission occurs (depopulation pumping).<sup>12</sup> In practice, optical pumping in  $^3\text{He}$  is

often done with a pressure of the order of 0.1 to 3 torr at room temperature, so that an intermediate situation occurs; an interpolation between the results corresponding to these two extreme cases will then be necessary.

A final simplification is obtained by considering the spontaneous emission from the  $2^3\text{P}$  level to the  $2^3\text{S}$  level as an instantaneous process; in other words, the lifetime  $\tau$  of the  $2^3\text{P}$  level is supposed to be zero (the real value is  $\tau \approx 10^{-7}$  s). In this way, the 18 populations of the  $2^3\text{P}$  level can be eliminated, and the only variables which remain are 6 populations for the metastable level, plus 2 Zeeman populations ( $n_+$  and  $n_-$ ) for the ground state. Clearly, our model is valid only if the pumping time  $\tau_p$  (inversely proportional to the laser power  $I$ ) is long compared to  $\tau$ , that is if stimulated emission from the  $2^3\text{P}$  level can be neglected, compared to spontaneous emission.

We shall not give here the details of the calculations which will be described more fully in Reference 60. The quantity of interest is the nuclear polarization  $M$  of the ground state as a function of the laser power  $I$  and of its frequency. One example of the results obtained is shown in Fig. 3; it has been assumed that the frequency of the laser is resonant with the  $2^3\text{S}_1 F=3/2 \leftrightarrow 2^3\text{P}_0$  transition (this corresponds to the  $C_9$  component of Figure 2). For this pumping transition, the values of  $M$  are found to be almost insensitive to collisional depolarization inside the  $2^3\text{P}$  level. After a linear increase, the curve of Figure 3 becomes sensitive to non linear effects which reduce its slope; although an infinite laser intensity would lead to a value of  $M$  very close to one, only  $M \approx 80\%$  is obtained if  $I = 5$  watts, which seems to be a reasonable maximum in practice (also it should be remembered that the model ignores stimulated emission and is not valid at very high laser intensities).

A better pumping efficiency is predicted by the model for the  $C_5$  component of Figure 2, when no collision depolarization is assumed inside the  $2^3\text{P}$  level. Unfortunately, even a small depolarization rate seems to reduce  $M$  very efficiently. In practice, for optical pumping experiments at room temperature, it is difficult to use gas pressures below 0.1 torr, and the  $C_8$  and  $C_9$  components seem to be the best choice for high nuclear polarizations.<sup>60</sup>

Figure 4 gives a general scheme of the experimental arrangement which was used to test these predictions. A pyrex cell, a few centimeters in diameter, is filled with  $^3\text{He}$  gas at a pressure ranging between 0.1 and 5 torr. A weak R.F. discharge in the gas populates the  $2^3\text{S}_1$  states (density  $\approx 10^{10}/\text{cc}$ ). The infrared light is provided by a colour-centre laser using  $(F_2^+)^*$  centres in NaF, developed by L. Mollenauer in 1980.<sup>61</sup> The NaF crystals are doped with  $\text{Mg}^{++}$  impurities (relative density  $\approx 10^{-4}$ ) acting as electron traps. The centres are created by electron bombardment (electron energy; 1.5 MeV; electron current:  $4.5 \mu\text{A}$ ; exposure time: 40 minutes; temperature; 80 K), followed by annealing at room temperature (24 hours). The crystals are normally stored at liquid nitrogen temperature although they can be kept at room temperature for a few days without significant loss of their laser properties. On the other hand, they have to be cooled to liquid nitrogen temperature during laser operation.

The colour centre laser is excited by a dye laser emitting in the infrared around the wavelength  $\lambda = 890 \text{ nm}$ ; the dye (HITC) is itself pumped by the red lines ( $\lambda = 644$  and  $690 \text{ nm}$ ) of a Kr<sup>+</sup> laser. A total power of 5 watts on these lines can provide 1 watt of infrared power, which in turn produces  $\approx 300 \text{ mW}$  of tunable light around  $\lambda = 1.08 \mu$ . The cavity of the colour centre laser is relatively long (1.5 m) and symmetrical (X shaped), with only one solid etalon inserted (thickness: 0.5 mm) in addition to a three plate birefringent Lyot filter. In this way, a



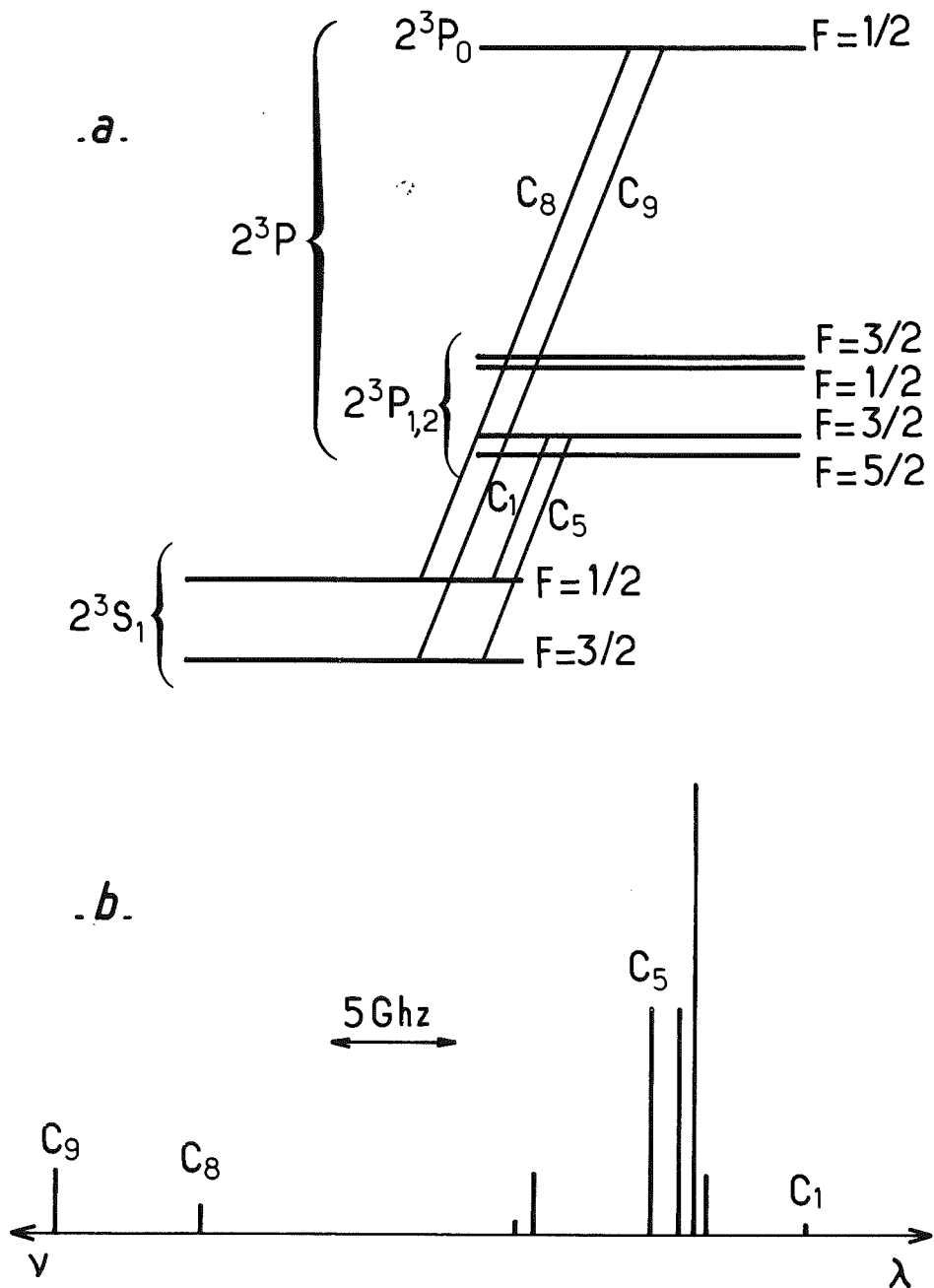


FIGURE 2 a. Level scheme of the  $2^3S$  and  $2^3P$  states of the  $^3\text{He}$  atom including fine and hyperfine structure. b. Components of the  $2^3S$ - $2^3P$  line ( $\lambda = 1.08 \mu$ ).

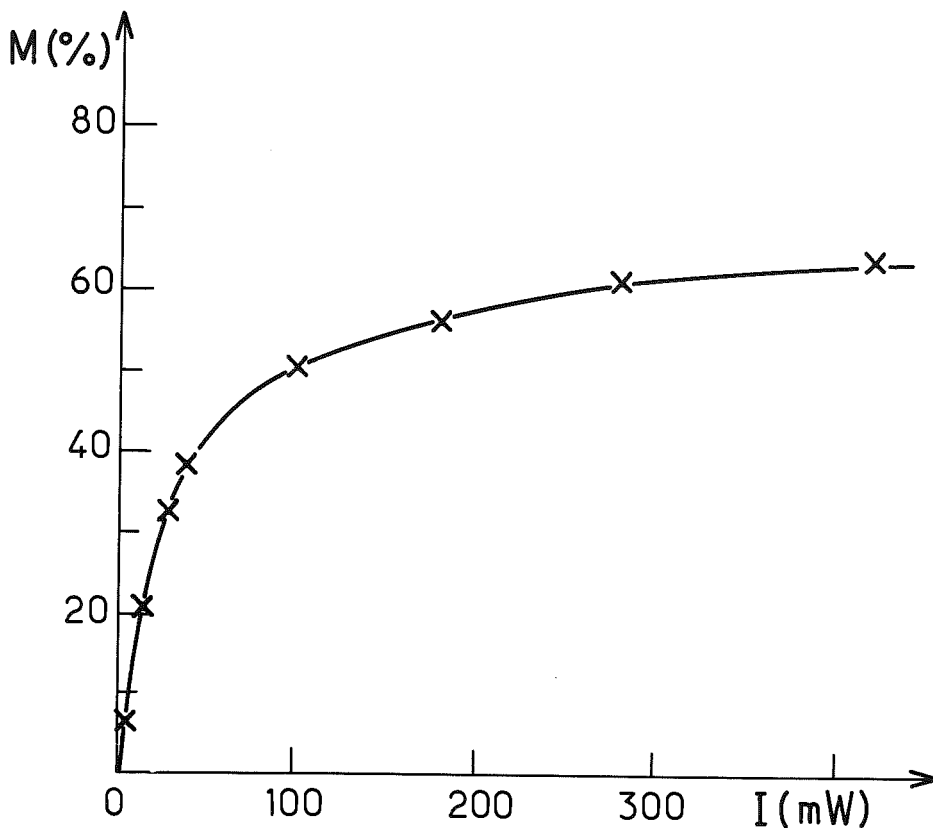


FIGURE 3 Nuclear polarization of the  $^3\text{He}$  gas as a function of laser light intensity. The crosses correspond to experimental results obtained with a multimode laser operating on the  $C_0$  component of the  $1.08 \mu$  line. The solid curve is the theoretical prediction obtained from a model discussed in the text.

few longitudinal modes ( $\approx 3$ ), which fall inside the Doppler width of the atomic absorption line, oscillate at the same time; this precaution avoids too much selection in the atomic velocity classes which are pumped and reduces saturation problems.

The pumping beam is sent through the  $^3\text{He}$  cell along the direction of the static magnetic field  $\mathbf{B}_0$ , and bounced on a mirror (not shown in Figure 4) so that it crosses the cell twice. This has two advantages: it increases the pumping intensity, and doubles the number of atomic velocity classes which are excited. The value of  $\mathbf{B}_0$  is not critical and a field of the order of 1 or 2 Gauss is convenient in practice.† Two methods can be used to measure  $M$ : a nuclear magnetic resonance method, as already used in the first experiments on  $^3\text{He}$  optical pumping,<sup>14</sup> or an optical detection method,<sup>62</sup> which is often more convenient to implement and gives a continuous reading of the nuclear polarization during its build-up. The principle of this optical method is to measure the degree of circular polarization of the light emitted by the helium gas discharge in the direction of the component of the

† Nevertheless, the homogeneity of the magnetic field over the cell is crucial. Magnetic gradients exceeding  $\approx 1$  mG/cm would create a relaxation time of  $M$  which would be shorter than  $\approx 1$  minute, and reduce dramatically the polarization obtained.

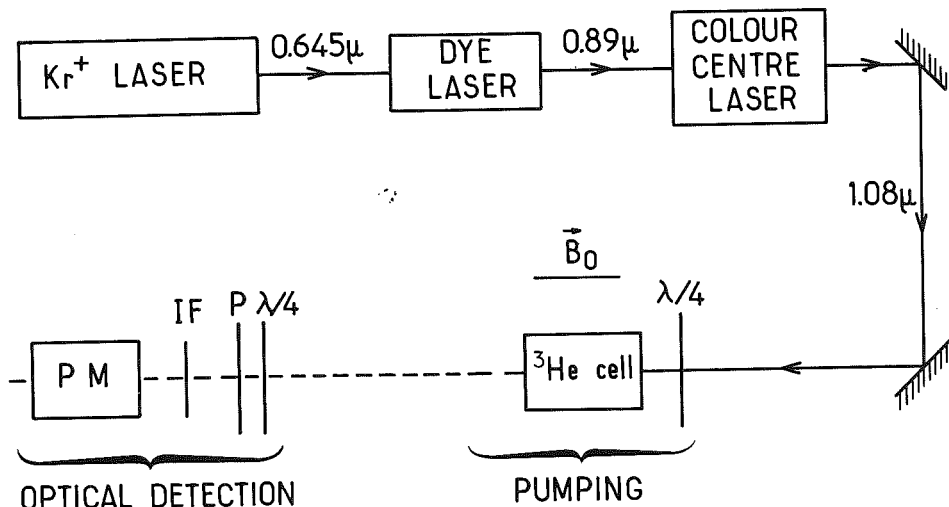


FIGURE 4 Experimental set-up of  $^3\text{He}$  optical pumping with a laser. The pumping light at  $1.08\ \mu$  is delivered by a colour centre laser excited by an infra-red dye laser. It is circularly polarized by a quarter wave plate ( $\lambda/4$ ) and sent onto the  $^3\text{He}$  cell along the magnetic field  $B_0$ . The optical detection of the polarization includes a rotating quarter wave plate  $\lambda/4$ , a linear polarizer  $P$ , an interference filter  $IF$  and a photomultiplier  $PM$ .

magnetization to be measured† [see Reference 63 for the calibration of this method].

How this is done in practice is schematically shown in Figure 4: the light emitted by the discharge crosses successively a rotating quarter-wave plate (rotation frequency  $\nu$ ), a fixed linear analyzer  $P$ , and an interference filter  $I.F.$  which selects the visible line  $\lambda = 668\ \text{nm}$  of helium. Left and right circular polarizations are alternatively detected by the photomultiplier, so that, when lock-in amplifier is used to select the modulated component at frequency  $2\nu$  of the photocurrent, one obtains an output signal which is directly proportional to the nuclear polarization of the gas.

The crosses in Figure 3 show the measured values of  $M$  as a function of the laser power  $I$  for the  $C_9$  component of the  $\lambda = 1.08\ \mu$  line ( $2^3S_1, F = 3/2 \leftrightarrow 2^3P_0$ ). The solid line is the result of the calculations using the model described above. The highest value of polarization obtained was  $M \approx 70\%$  with a laser power  $I$  of 300 mW centered on the  $C_8$  component ( $2^3S_1, F = 1/2 \leftrightarrow 2^3P_0$ ) and a cell filled at a pressure  $p = 0.3$  torr of  $^3\text{He}$ . The pressure dependence of the nuclear polarization obtained by this method is shown in Figure 5. At low pressures ( $p < 0.3$  torr), the fast diffusion of the metastable atoms towards the walls of the container limits their lifetime and therefore their number density; in addition, very weak discharges are difficult to maintain in a low pressure gas and optical pumping is not optimum. At high pressures ( $p \geq 2$  torr), other physical processes (metastable-metastable ionizing collisions, etc. . .) limit the number density of metastable atoms, which does not increase linearly as a function of  $p$  as for the ground state atoms; consequently, the proportion of pumped atoms becomes insufficient and high values of  $M$  are not obtained.

† When an atom is nucleary-oriented in the ground state, it conserves its orientation during the excitation processes (e.g. electron collisions) which bring it to various excited states. During the lifetime  $\tau$  of the excited state, the hyperfine coupling  $a\mathbf{I} \cdot \mathbf{J}$  transfers part of the orientation of  $\mathbf{I}$  to  $\mathbf{J}$  (the larger  $\tau$  and  $a$ , the more efficient this process). Subsequently, when the atom emits a photon and returns to a lower state, this photon is (partially) circularly polarized.

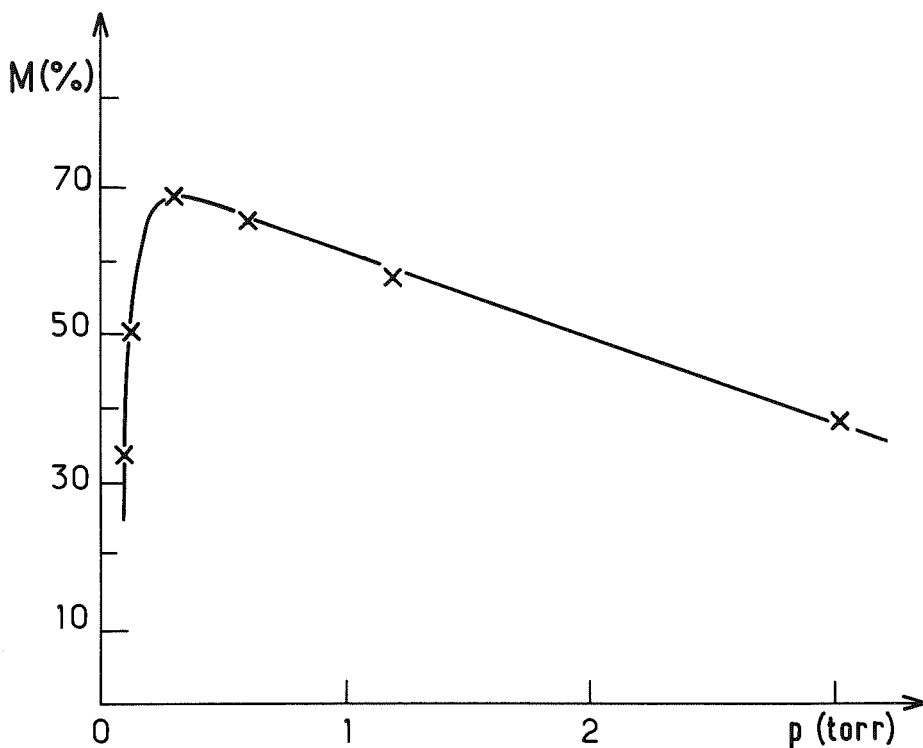


FIGURE 5 Nuclear polarization obtained in a  $^3\text{He}$  gas by optical pumping as a function of the  $^3\text{He}$  pressure.

Similar experiments were repeated at low temperatures (4.2 K and below), using pyrex cells internally coated with solid hydrogen films in order to inhibit the very fast nuclear relaxation which would be introduced by very cold bare pyrex walls.<sup>8,65</sup> At these low temperatures, the model described above fails to accurately predict the values of  $M$  because the metastability exchange collisions are no longer an efficient process to couple the orientation of the metastable and ground state levels: the cross-section of such collisions decreases by more than two orders of magnitude between 300 K and 4.2 K,<sup>66-68</sup> because of the repulsive barrier in the interaction potential between the  $2^3\text{S}_1$  and  $1^1\text{S}_0$  helium atoms.<sup>69,70</sup> Other unknown processes (e.g. metastable deexcitation on the walls) might become more efficient to transfer the polarization to the ground state at low temperatures. Experimentally, it was found that values of  $M$  exceeding 25% are not obtained by optical pumping at 4.2 K;  $M$  tends to saturate when the laser power is increased above 20 mW. We describe in the next section another polarization technique which does not suffer from the same limitations.

#### A POLARIZATION TRANSFER TECHNIQUE TO POLARIZE GASEOUS $^3\text{He}$ AT LOW TEMPERATURES

It is possible to optically pump a  $^3\text{He}$  gas at room temperature, as described above, and to transport the nuclear polarization by spin diffusion to a gas at low temperatures. This method requires the use of two cells at different temperatures,

coupled by a narrow tube in which spin diffusion occurs. A sketch of such an experiment is shown in Figure 6. The larger cell, of volume  $V_1$ , is at room temperature  $\theta_1$ ;  $V_1$  is of the order of 100 cc and is chosen for optimum optical pumping at room temperature. The smaller cell, of volume  $V_2$ , is immersed in a liquid helium bath, so that its temperature  $\theta_2$  can be varied between 4.2 and

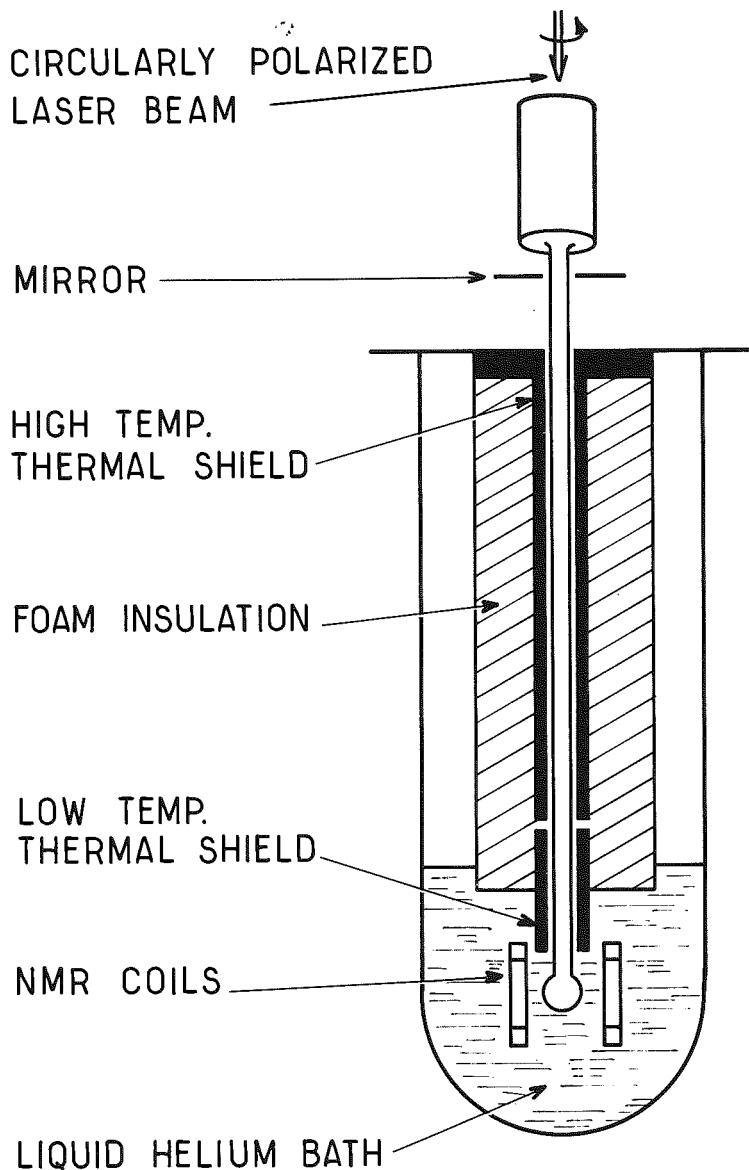


FIGURE 6 Sketch of the double cell experiment for polarizing  $^3\text{He}$  at low temperatures. The upper cell at 300 K is submitted to optical pumping. The nuclear polarization is transferred by diffusion to the lower cell immersed in a liquid helium bath. The thermal shields and foam insulation are used to control the temperature gradients along the tube connecting the two cells, and to minimize the relaxation of the nuclear spins on the walls at intermediate temperatures.

1.5 K; typical values of  $V_2$  range from 1 to 10 cc. The tube is 70 cm long and its internal diameter is small enough so that its volume is negligible compared to  $V_1$  and  $V_2$ . A weak discharge is sustained in the upper cell. The laser light is directed on the upper cell (Figure 6), parallel to the vertical magnetic field  $\mathbf{B}_0$ . This field is provided by a set of 3 (or 5) coils centered on the same axis and whose relative positions and currents are carefully adjusted so as to keep a good homogeneity ( $\approx 10^{-4}$ ) of  $\mathbf{B}_0$  all along the tube connecting  $V_1$  and  $V_2$ .

The amount of  $^3\text{He}$  is chosen so that the density  $n_1$  in  $V_1$  is optimum for a weak discharge and efficient optical pumping ( $n_1 \approx 10^{16} - 10^{17} \text{ cm}^{-3}$ , as seen in the above section). Consequently, the density  $n_2$  in  $V_2$  is much larger than  $n_1$  (since the diameter of the tube is large compared to the mean free path in the gas, the pressures equilibrate so that  $n_1\theta_1 = n_2\theta_2$ ). For instance, at  $\theta_2 = 3 \text{ K}$ , one reaches  $n_2 \approx 10^{18}/\text{cc}$ . Some molecular hydrogen is added to the  $^3\text{He}$  gas, and freezes on the cold parts of the inside wall of the cell; this ensures that no fast nuclear relaxation occurs.<sup>8</sup> Enough hydrogen must be included so that the hydrogen film is at least 5 layers thick in the lower cell. Also, some care must be taken in controlling the temperature gradient along the connecting tube, so as to obtain a good transfer of polarization to  $V_2$ . This is the reason for the two thermal shields shown in Figure 6; one of them fixes the temperature of the lower part of the tube close to  $\theta_2$ , the other keeps the temperature of the rest of the tube to a value near 100 K. The gap between the two shields is small; this minimizes the region of strong temperature gradient where  $^3\text{He}$  atoms might relax rapidly on pyrex surfaces (cold enough to strongly adsorb  $^3\text{He}$  but too warm to be coated with a sufficient amount of molecular hydrogen).

In this geometry, it is justified to assume well-defined average magnetizations  $M_1$  and  $M_2$  respectively in the pumping cell  $V_1$  and in the bottom cell  $V_2$  (the diffusion time for an atom to cross either cell is very much shorter than the average time for an atom to diffuse from one cell to another). It is convenient in this kind of experiment to constantly monitor  $M_1$  by the optical detection technique mentioned above.<sup>62</sup> On the other hand, no discharge is sustained in the lower cell, and the gas density is high; for both these reasons, a NMR measurement of  $M_2$  is more desirable.

To give a specific example, let us consider a double cell of volumes  $V_1 = 85 \text{ cm}^3$ ,  $V_2 = 10 \text{ cm}^3$ , filled at room temperature with 5 torr of  $^3\text{He}$  and 0.1 torr of  $\text{H}_2$ . If  $\theta_2 = 4.2 \text{ K}$ , this corresponds to a  $^3\text{He}$  pressure of  $\approx 0.5$  torr, that is number densities  $n_2 \approx 1.2 \cdot 10^{18} \text{ cm}^{-3}$  and  $n_1 \approx 1.7 \cdot 10^{16} \text{ cm}^{-3}$  ( $n_1$  is therefore the right order of magnitude for efficient optical pumping). When the upper cell is continuously pumped,  $M_2$  increases slowly and reaches a maximum after a delay of the order of 1 hour. Experimentally, the transfer of polarization between  $V_1$  and  $V_2$  was found to be 100% within the error bars of the measurement ( $\pm 10\%$ ). Values of  $M_2$  reaching 50% were obtained at 4.2 K. The density of magnetization  $n_2 M_2$  was of the order of  $6 \times 10^{17} \text{ cm}^{-3}$ , that is comparable (with higher  $M_2$  but lower  $n_2$  values) to what has been achieved by compressing polarized gas at room temperature with a Toepler pump.<sup>44,45</sup> The lifetime of  $M_2$  was of the order of a few hours and presumably limited by spin diffusion towards regions with faster relaxation at higher temperatures (the relaxation time of  $^3\text{He}$  nuclear spins on solid  $\text{H}_2$  can be as long as days).<sup>64</sup>

When the lower cell is polarized, it is possible to cool it down to  $\theta_2 = 1.5 \text{ K}$  in about 5 minutes without substantial loss of polarization (1.5 K is roughly the lowest temperature reachable with our equipment). Below 2 K, the lifetime of the polarization was found to be of the order of 20 minutes and only weakly

temperature dependent. The polarization transfer technique can also be directly used when the lower cell is at temperatures  $\theta_2$  below 4.2 K. Actually, the method seems to work slightly better if  $\theta_2 \approx 3$  K than 4.2 K, mainly because of the reduction of the partial pressure of  $H_2$  (the efficiency of optical pumping is strongly reduced if the  $2^3S$  metastable atoms are quenched by collisions against  $H_2$  molecules). No systematic study has been made yet of the efficiency of the polarization transfer technique as a function of  $\theta_2$ .

Several possibilities could be tried to increase the density of the polarized gas. As shown in Figure 5, the filling pressure of the cells could be multiplied by a factor 10 without too much decrease in the efficiency of optical pumping. Another possibility to obtain denser polarized gas would be to use lower temperatures; recent experiments<sup>9</sup> have shown that super-fluid  $^4He$  films can be very efficient to reduce the nuclear relaxation on the walls down to 500 mK, and probably at even lower temperatures.

In the appendix of this article, we describe a simple model to understand the time evolution of the magnetization in the two cells, as well as their limiting values.

## CONCLUSION

Optical pumping in  $^3He$  and  $^4He$  is a technique which seems to offer many potential applications, ranging from the production of polarized beams or polarized targets to the study of hot plasmas or clean surfaces in solid state physics. Some progress has been made recently, due to the use of lasers as pumping sources and low temperature techniques. Further improvements could probably be obtained in the future by using laser diode arrays which, in some cases,<sup>71</sup> can produce high c.w. powers (several watts); they could for example be used to pump colour centre lasers. Another improvement would be to further compress the polarized gas at low temperature, as already done at room temperature,<sup>44,45</sup> in order to increase the density. Similar techniques have been recently used with success in the case of spin-polarized hydrogen.<sup>72,73</sup> Even higher densities might be obtained by polarizing a liquid droplet by atom exchange with a spin polarized vapour,<sup>58</sup> which would open up the field of the study of degenerate polarized liquids.

## APPENDIX

### RATE EQUATIONS FOR A DOUBLE CELL

In order to better understand the build-up of the magnetization  $M_2$  in the lower cell, we develop here a simple model to describe the coupling between  $M_1$  and  $M_2$  and their evolution in time. In the upper cell, it is a good approximation to assume that light coupling between the polarization of the ground and metastable levels leads to a single rate for the pumping of the magnetization  $M_1$ . Let us first consider the simple case (Figure 7a) where the volume of the connecting tube is completely negligible, compared to  $V_1$  and  $V_2$  and where there is no loss of

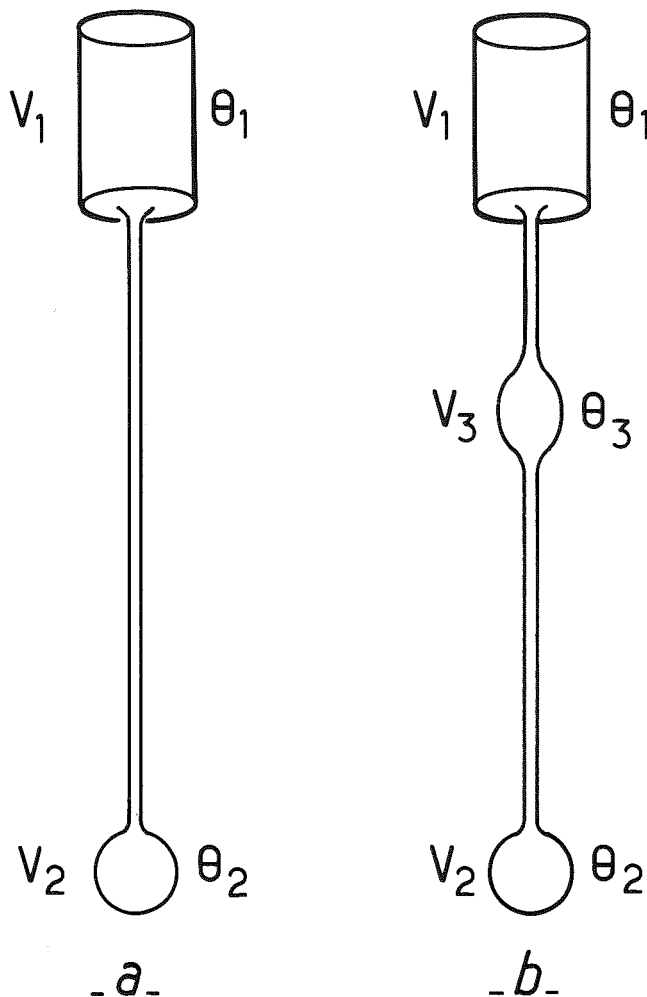


FIGURE 7 Models for understanding the coupling of orientations in the double cell experiment. a) One assumes a volume  $V_1$  at temperature  $\theta_1$  is connected with a volume  $V_2$  at temperature  $\theta_2$  by a tube of negligible volume. b) In addition, one assumes a non-zero volume  $V_3$  at an intermediate temperature  $\theta_3$  where some relaxation takes place during diffusion between the upper cell  $V_1$  and the lower cell  $V_2$ .

magnetization in the connecting tube. The equations of evolution of  $M_1$  and  $M_2$  are then:

$$\begin{aligned} \dot{M}_1 &= \gamma_p(M_0 - M_1) + cx(M_2 - M_1) - \Gamma_1 M_1 \\ \dot{M}_2 &= c(M_1 - M_2) - \Gamma_2 M_2 \end{aligned} \quad (1)$$

where  $c$  is the inverse of the average time for an atom within  $V_2$  to diffuse to  $V_1$ , and  $x$  is the ratio of the number  $N_2$  of atoms in  $V_2$  to the number  $N_1$  in  $V_1$  ( $x = N_2/N_1 = V_2\theta_1/V_1\theta_2$ );  $\Gamma_1$  and  $\Gamma_2$  are the average rates at which magnetization is destroyed within cells  $V_1$  and  $V_2$ , respectively;  $M_0$  is the magnetization to which the pumping cell would be pumped if  $c$  and  $\Gamma_1$  were zero, and  $\gamma_p$  is the pumping rate.



Equations (1) provide a very simple description of the magnetization coupling in the absence of magnetization loss in the connecting tube. In order to take some account of losses in the tube, consider the relatively simple extension of this picture to the case where there is a single relaxing region somewhere in the tube having volume  $V_3$ , as shown in Figure 7b, where there is a well-defined magnetization  $M_3$  coupled to  $M_1$  and  $M_2$  by diffusion. In that case, the equations become:

$$\begin{aligned} \dot{M}_1 &= \gamma_p(M_0 - M_1) + c_1 \frac{N_3}{N_1} (M_2 - M_1) - \Gamma_1 M_1 \\ \dot{M}_3 &= c_1(M_1 - M_3) + c_2(M_2 - M_3) - \Gamma_3 M_3 \\ \dot{M}_2 &= c_2 \frac{N_3}{N_2} (M_3 - M_2) - \Gamma_2 M_2 \end{aligned} \quad (2)$$

where  $c_1$  and  $c_2$  are the inverse of the time for an atom in  $V_3$  to diffuse to  $V_1$  and  $V_2$  respectively. As long as the number of atoms in volume  $V_3$  is very much less than the numbers in  $V_1$  and  $V_2$ ,  $M_3$  relaxes to a quasi-equilibrium value:

$$M_3 \approx \frac{c_1 M_1 + c_2 M_2}{c_1 + c_2 + \Gamma_3} \quad (3)$$

on a time scale that is very short compared to the time for an appreciable change of magnetization in  $V_1$  or  $V_2$ . On the time scale in which we are interested, we can then substitute Equations (3) in Equations (2) to obtain:

$$\begin{aligned} \dot{M}_1 &= \gamma_p(M_0 - M_1) + cx(\beta M_2 - M_1) - \Gamma_1 M_1 \\ \dot{M}_2 &= c(\alpha M_1 - M_2) - \Gamma_2 M_2 \end{aligned} \quad (4)$$

where the coefficient  $c$  is now given by:

$$c = \frac{c_2(c_1 + \Gamma_3)N_3}{(c_1 + c_2 + \Gamma_3)N_2}$$

This coefficient plays the role of an effective rate of transfer of magnetization from one cell to another, subject to loss probabilities  $\alpha = c_1/(c_1 + \Gamma_3)$  on the way from  $V_1$  to  $V_2$ , and  $\beta = c_2/(c_2 + \Gamma_3)$  on the way from  $V_2$  to  $V_1$ . The coefficient  $x$  is now:

$$x = \frac{\alpha V_2 \theta_1}{\beta V_1 \theta_2}$$

Comparing Equations (4) to Equations (1), it seems reasonable to take account of some loss of magnetization during diffusion through the connecting tube by simply introducing phenomenological  $\alpha$  and  $\beta$  into Equations (1) to get Equations (4), provided that the loss of magnetization during transfer is not large. When  $M_{10}$  and  $M_{20}$  are the initial polarizations at time  $t = 0$ , the solutions of Equations (4) can be written:

$$\begin{aligned} M_1 &= M_{1e} + \left[ \frac{f - \gamma_2}{f - s} (M_{10} - M_{1e}) - \frac{\beta cx}{f - s} (M_{20} - M_{2e}) \right] e^{-ft} \\ &\quad + \left[ \frac{\gamma_2 - s}{f - s} (M_{10} - M_{1e}) + \frac{\beta cx}{f - s} (M_{20} - M_{2e}) \right] e^{-st} \\ M_2 &= M_{2e} + \left[ \frac{\gamma_2 - s}{f - s} (M_{20} - M_{2e}) - \frac{c\alpha}{f - s} (M_{10} - M_{1e}) \right] e^{-ft} \\ &\quad + \left[ \frac{f - \gamma_2}{f - s} (M_{20} - M_{2e}) + \frac{c\alpha}{f - s} (M_{10} - M_{1e}) \right] e^{-st} \end{aligned} \quad (5-a)$$

with:

$$\begin{aligned}\gamma_1 &= \gamma_p + cx + \Gamma_1 \\ \gamma_2 &= c + \Gamma_2\end{aligned}\quad (5-b)$$

$$M_{2e} = \frac{c\alpha}{\gamma_2} M_{1e}$$

and

$$M_{1e} = \frac{\gamma_2 \gamma_p M_0}{\gamma_1 \gamma_2 - c^2 \alpha \beta x} \quad (5-c)$$

The magnetizations go exponentially to their equilibrium values  $M_{2e}$  and  $M_{1e}$  with the fast and slow recovery rates:

$$f = \frac{1}{2}\gamma_1 + \frac{1}{2}\gamma_2 + \frac{1}{2}\sqrt{(\gamma_1 - \gamma_2)^2 + 4c^2\alpha\beta x} \quad (6)$$

$$s = \frac{1}{2}\gamma_1 + \frac{1}{2}\gamma_2 - \frac{1}{2}\sqrt{(\gamma_1 - \gamma_2)^2 + 4c^2\alpha\beta x}$$

Equation (4) and their solutions (5) hold whether or not there is any pumping and for any initial conditions. For example, for pumping starting from zero magnetization, they simplify to:

$$\begin{aligned}M_1 &= M_{1e} [1 - (1 - \Delta)e^{-ft} - \Delta e^{-st}] \\ M_2 &= M_{2e} \left[ 1 - \frac{f}{f-s} e^{-st} + \frac{s}{f-s} e^{-ft} \right]\end{aligned}\quad (7)$$

with  $M_{1e}$ ,  $M_{2e}$ ,  $f$ , and  $s$  given as above and:

$$\Delta = \frac{(\gamma_2 - s)(\gamma_1 + \gamma_2 - s)}{\gamma_2(f - s)} \quad (8)$$

Alternatively, when the magnetizations have been pumped to equilibrium and the pumping then stopped, the decay of the magnetizations to zero is given by:

$$\begin{aligned}M_1 &= M_{1e} [(1 - \Delta)e^{-ft} + \Delta e^{-st}] \\ M_2 &= M_{2e} \left[ \frac{f}{f-s} e^{-st} - \frac{s}{f-s} e^{-ft} \right]\end{aligned}\quad (9)$$

where  $M_{1e}$  and  $M_{2e}$  are given by Equations (5-c) with  $\gamma_p \neq 0$  [pumping still going on], while the other constants  $f$ ,  $s$  and  $\Delta$  are obtained from (6) and (8) with  $\gamma_p = 0$ .

Figure 8 shows an example of the decay of  $M_1$  from equilibrium magnetization, as observed optically on the upper cell (the temperature  $\theta_2$  was 4.2 K). The data are fit nicely by the sum of two exponentials:

$$M_1 = M_{1e} (0.27e^{-0.82t} + 0.73e^{-0.018t}) \quad (10)$$

which, using Equations (6) and (8), gives:

$$\begin{aligned}\gamma_1 &= 0.013 \text{ s}^{-1} & \gamma_2 &= 0.001 \text{ s}^{-1} \\ xc^2\alpha\beta &= 9.4 \times 10^{-6}\end{aligned}$$

The  $\gamma_2$  value is to be compared to the relaxation probability  $\Gamma_2$  (relaxations are long at 4.2 K, of the order of several hours). Since  $\Gamma_2 \ll \gamma_2$ :

$$\gamma_2 \approx c$$

which means that the time constant of evolution of the bottom polarization  $M_2$  is

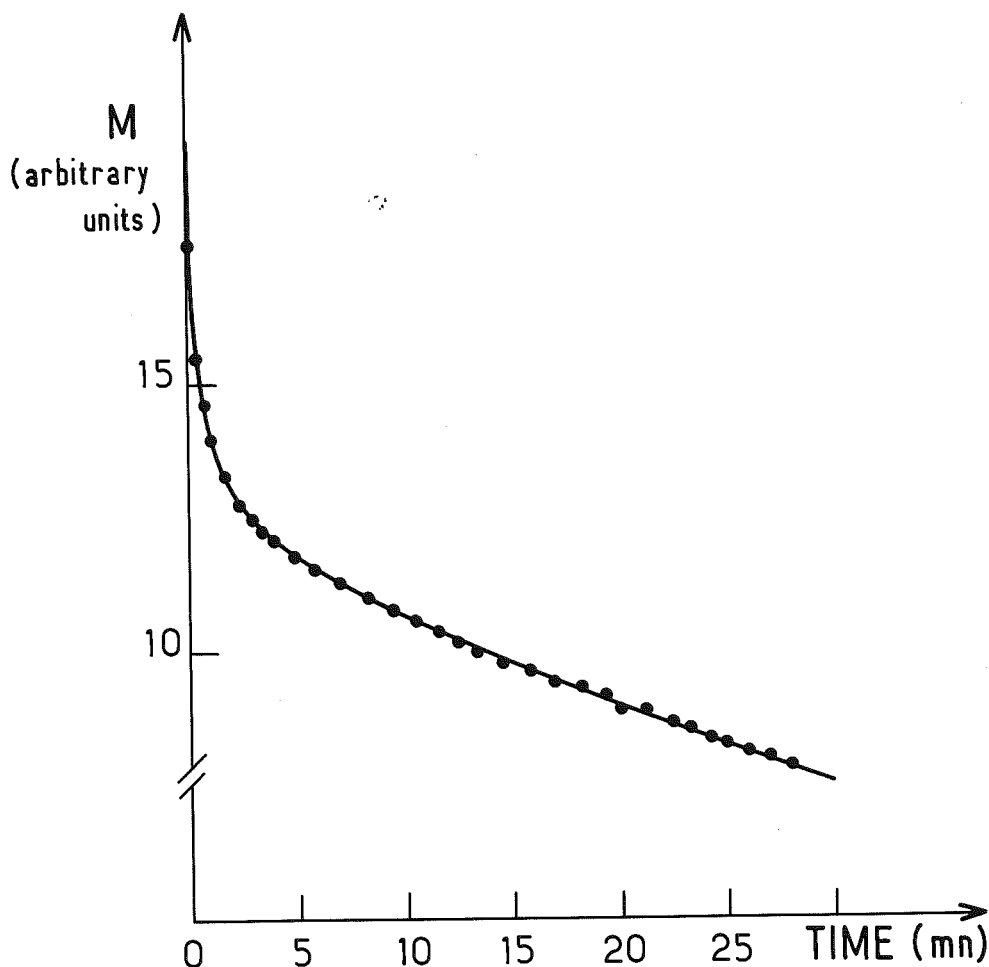


FIGURE 8 Experimental result obtained in the double cell experiment. After an equilibrium is reached for nuclear polarization between the upper and the lower cell, the optical pumping is stopped and one then monitors the decrease of polarization in the upper cell; it then decays with two different time constants.

primarily governed by the coupling with the upper cell. Thus, from (5-b), the ratio of equilibrium magnetization  $M_{2e}/M_{1e}$  simplifies to  $\alpha$ . The value of  $\alpha$  can be derived from the measured quantity:

$$xc^2 \cdot \alpha\beta = \alpha^2 \times \frac{N_2}{N_1} \times \gamma_2^2 = 9.4 \cdot 10^{-6}.$$

With:

$$\frac{N_2}{N_1} = 9.3 \quad \text{and} \quad \gamma_2 = 10^{-3} \text{ s}^{-1},$$

one obtains:

$$\alpha = 0.96.$$

The uncertainty on this number depends primarily on the number densities. One finds from the decay curve of Figure 8 and the above rate equations that the

magnetization transfer between the upper cell and the low cell should be of the order of 96%.

### References

- 1 See, for example: Keller, W. E. *Helium-3 and Helium-4*, Plenum Press, New York (1969).
- 2 Castaing, B. and Nozières, P. *J. Physique*, **40**, 257 (1979).
- 3 "Spin Polarized Quantum Systems", *J. Physique*, Coll, **41**, C-7 (1980), and references therein.
- 4 Pasell, L. and Schermer, R. I. *Phys. Rev.* **150**, 146 (1966).
- 5 Richards, M. G. "Nuclear Magnetic Resonance in Adsorbed Helium", in "Phase Transitions in Surface Films", edited by Dash, J. G. and Ruvalds, J. Plenum Press (1980).
- 6 Chapellier, M. *J. Physique Lett.* **43**, L-609 (1982).
- 7 Hammel, P. C., Roukes, M. L., Hu, Y., Gramila, T. J., Mamiya, T., and Richardson, R. C. *Phys. Rev. Lett.* **51**, 2124 (1983).
- 8 Barbé, R., Laloë, F. and Brossel, J. *Phys. Rev. Lett.* **34**, 1488 (1975).
- 9 Himbert, M., Lefevre-Seguin, V., Nacher, P. J., Dupont-Roc, J., Leduc, M. and Laloë, F. *J. Physique Lett.* **44**, L-523 (1983).
- 10 Walters, G. K. Proceedings of the International Conference on Polarized Targets and Ion Sources in Saclay (France). P. 201 (1966). Edited by C. E. N. Saclay, B. P. n° 2. 91191 Gif-sur-Yvette Cedex, France.
- 11 Cohen-Tannoudji C. et Kastler, A. Progress in Optics. vol. V (1966). North Holland Publ. Co.
- 12 Happer, W. *Rev. Mod. Phys.* **44**, 169 (1972).
- 13 Bouchiat, M. A., Carver, T. R. and Varnum, C. M. *Phys. Rev. Lett.* **5**, 373 (1960).
- 14 Colegrove, F. D., Schearer, L. D. and Walters, G. K. *Phys. Rev.* **132**, 2561 (1963).
- 15 Daniels, J. M. and Timsit, R. S. *Canad. J. Phys.* **49**, 525, 539 and 545 (1971).
- 16 Bley, P. and Turowski, P. *Z. Angew. Phys.* **24**, 57 (1968).
- 17 Beckmann, R., Holm, U. and Lindner, B. *Z. Phys. A* **275**, 319 (1975).
- 18 Dupont-Roc, J., Leduc, M. et Laloë, F. *J. Physique* **34**, 961 and 977 (1973).
- 19 Nacher, P. J., Leduc, M., Trenec, G. et Laloë, F. *J. Physique Lett.* **43**, L-525 (1982).
- 20 Leduc, M., Nacher, P. J., Crampton, S. B. and Laloë, F. Proceedings of the "Quantum Fluids and Solids" Conference. (Sanibel, USA, April 1983).
- 21 Lhuillier C. et Laloë, F. *J. Physique* **40**, 239 (1979).
- 22 Lhuillier C. et Laloë, F. *J. Physique* **43**, 197 and 225 (1982).
- 23 Baker, S. D., Carter, E. B., Findley, D. O., Hatfield, L. L., Phillips, G. C., Stockwell, N. D. and Walters, G. K. *Phys. Rev. Lett.* **20**, 738 (1968).
- 24 Leduc, M. and Laloë, F. *Opt. Comm.* **3**, 56 (1971); Pinard, M. and Leduc, M. *J. Physique* **35**, 741 (1974).
- 25 McCusker, M. V., Hatfield, L. L. and Walters, G. K. *Phys. Rev. Lett.* **22**, 817 (1969); *Phys. Rev. A* **5**, 177 (1972).
- 26 Keliher, P. J., Gleason, R. E. and Walters, G. K. *Phys. Rev. A* **11**, 1279 (1975).
- 27 Schearer, L. D. and Riseberg, L. A. *Phys. Lett.* **33A**, 325 (1970).
- 28 Schearer, L. D. *Ph.D. Thesis*, Rice University (1966).
- 29 Byerly, H. R. *Ph.D. Thesis*, Rice University (1967).
- 30 Wigner, E. *Nachr. Akad. Wiss., Göttingen. Math.-Physik Chem. Abt.* **1927**, 375.
- 31 Schmeltekopf, A. C. and Broida, H. P. *J. Chem. Phys.* **39**, 1261 (1963).
- 32 Schearer, L. D. *Phys. Rev. Lett.* **22**, 629 (1969); Schearer, L. D. and Holton, W. C. *Phys. Rev. Lett.* **24**, 1214 (1970).
- 33 Schearer, L. D. *Phys. Lett.* **31A**, 457 (1970).
- 34 Schearer, L. D. and Riseberg, I. A. *Phys. Lett.* **35A**, 267 (1971).
- 35 Schearer, L. D. *Phys. Rev.* **A10**, 1380 (1974).
- 36 Fahey, D. W., Parks, W. F. and Schearer, L. D. *J. Phys. B* **12**, L-619 (1979); *Phys. Rev.* **A20**, 1372 (1979).
- 37 Hamel, J. C. R. *Acad. Sci. (Paris)*, **277**, B-523 (1973).
- 38 Hamel, J. and Vienne, J. F. *Opt. Comm.* **7**, 83 (1973); Hamel, J. and Barrat, J. P. *Opt. Comm.* **10**, 331 (1974); Hamel, J., Margerie, J. and Barrat, J. P. *Opt. Comm.* **12**, 409 (1974); David, B. Hamel, J. and Barrat, J. P. *Opt. Comm.* **32**, 241 (1980).
- 39 Dmitriev, S. P., Zhitnikov, R. A. and Okunevich, A. I. *Zh. Eksp. Teor. Fiz.* **70**, 69 (1976); *Sov. Phys. JETP* **43**, 35 (1976).
- 40 Zhitnikov, R. A. page 308 Proceedings of the 6th Intern. Conf. on Atomic Physics, Riga (USSR), 1978. Dmitriev, S. P., Zhitnikov, R. A., Kartoshkin, V. A., Klementev, G. V. and Melnikov, V. D. *Zh. Eks. Teor. Fiz.* **85**, 840 (1983).

- 41 Hodge, L. A., Dunning, F. B. and Walters, G. K. *Rev. Sci. Instr.* **50**, 1 (1979).
- 42 Gray, L. G., Giberson, K. W., Chu Cheng, Keiffer, R. S., Dunning, F. B. and Walters, G. K. *Rev. Sci. Instr.* **54**, 271 (1983).
- 43 Phillips, G. C., Perry, R. R., Windham, P. M., Walters, G. K., Schearer, L. D. and Colegrove, F. D. *Phys. Rev. Lett.* **9**, 502 (1962).
- 44 Timsit, R. S., Daniels, J. M., Dennig, E. I., Kiang, A. K. C. and May, A. D. *Canad. J. Phys.* **49**, 508 (1971).
- 45 Timsit, R. S., Hilger, W. and Daniels, J. M. *Rev. Sci. Instr.* **44**, 1722 (1973).
- 46 McCamis, R. H., Verheijen, P. J. T., Maughan, G., Okumusoglo, N. T., van Oers, W. T. H., Daniels, J. M. and May, A. D. AIP Conference Proceedings n° 69. "Polarization Phenomena in Nuclear Physics 1980", Fifth International Symposium, Santa Fe. Verheijen, P. J. T., McCamis, R. H., Lapointe, C. and Van Oers, W. T. H. Communication to the Few Body Conference X". (Karlsruhe, RFA, August 1983).
- 47 Pasell, L. private communication.
- 48 Williams, W. G. *Nukleonika*, **25**, 769 (1980).
- 49 Freeman, F. F. and Williams, W. G. *J. Phys. E* **11**, 459 (1978).
- 50 Golub, R. *J. Physique Lett.* **44**, L-321 (1983).
- 51 Physics Today, Aug. 1982, p. 17.
- 52 Kulsrud, R. M., Furth, H. P., Valeo, E. J. and Goldhaber, M. *Phys. Rev. Lett.* **49**, 1248 (1982).
- 53 More, R. M. *Phys. Rev. Lett.* **51**, 396 (1983).
- 54 Blue, T. E., Blue, J. W., Durham, J. S., Harris, D. B., Hnesh, A. S. and Reyes, J. J. *J. Appl. Phys.* **54**, 615 (1983).
- 55 Murnick, D. E. *Appl. Phys. Lett.* **42**, 544 (1983).
- 56 Riddle, T. W., Onellion, M., Dunning, F. B. and Walters, G. K. *Rev. Sci. Instr.* **52**, 797 (1981).
- 57 Onellion, M., Hart, M. W., Dunning, F. B. and Walters, G. K. *Phys. Rev. Lett.* **52**, 380 (1984).
- 58 McAdams, H. H. and Walters, G. K. *Phys. Rev. Lett.* **18**, 436 (1967). McAdams, H. H. *Phys. Rev.* **170**, 276 (1968).
- 59 Partridge, R. B. and Series, G. W. *Proc. Phys. Soc.* **88**, 983 (1966).
- 60 Nacher, P. J. et Leduc, M. In preparation.
- 61 Mollenauer, L. F. *Opt. Lett.* **5**, 188 (1980).
- 62 Pavlović, M. and Laloë, F. *J. Physique* **31**, 173 (1970).
- 63 Pinard, M. These de 3ème cycle. Univ. Paris VI (1973), see also next reference.
- 64 Pinard, M. and van der Linde, J. *Canad. J. Phys.* **52**, 1615 (1974).
- 65 Lefevre-Seguín, V. Thèse, Paris (1984); Lefevre-Seguín, V., Nacher, P. J., Brossel, J. Hardy, W. N. and Laloë, F. "Relaxation nucléaire de  $^3\text{He}$  sur  $\text{H}_2$  solide", in préparation.
- 66 Colegrove, F. D. Schearer, L. D. and Walters, G. K. *Phys. Rev.* **135A**, 353 (1964).
- 67 Rosner, S. D. and Pipkin, F. M. *Phys. Rev.* **A5**, 1909 (1972).
- 68 Barbé, R. *J. Phys.* **B9**, 995 (1976).
- 69 Kolker, H. J. and Michels, H. H. *J. Chem. Phys.* **50**, 1762 (1969).
- 70 Hickman, A. P. and Lane, N. F. *Phys. Rev.* **A10**, 444 (1974).
- 71 Scifres, D. R., Burnham, R. D. and Streifer, W. *Appl. Phys. Lett.* **41**, 118 (1982).
- 72 Sprik, R., Walraven, J. T. M. and Silvera, I. F. *Phys. Rev. Lett.* **51**, 479 (1983).
- 73 Hess, H. F., Bell, D. A., Kochanski, G. P., Cline, R. W., Kleppner, D. and Greytak, T. J. *Phys. Rev. Lett.* **51**, 483 (1983).