Optical polarization of helium-3 nuclei

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The present state of the problem of producing highly polarized systems of helium-3 by laser optical pumping over a broad temperature range is reviewed. The physical principles underlying the polarization of ^{3}He nuclei during optical pumping and the exchange of metastability are described. Particular features of laser pumping at low temperatures are discussed. The possible use of polarized helium atoms in research on exchange and relaxation processes, in quantum magnetometry, and in nuclear physics to produce polarized targets and particle beams is discussed. The results of theoretical and experimental research on the quantum properties of highly polarized systems at low temperatures, near the temperature of quantum degeneracy, are reviewed.

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1. INTRODUCTION

Nuclei of the ^{3}He isotope in the liquid or gaseous state can be polarized by several methods. The simplest, at least in principle, is the "brute-force" method of immersing the sample in a strong magnetic field and then cooling it. This method is rather inefficient, however: Simple calculations show that at a temperature ~ 1 K in a magnetic field of the order 10 T (10^5G) an equilibrium degree of nuclear polarization of ^{3}He no greater than $P_n \sim 1\%$ can be achieved; even if the temperature is lowered to ~ 0.2 K, it is still not possible to surpass $P_n \sim 4\%$. In practice, the efficiency of this method is even lower, since at $T < 0.4$ K liquid ^{3}He is a degenerate Fermi system with a constant and extremely small magnetic susceptibility.[1] Polarizing ^{3}He nuclei by imposing a strong magnetic field is reasonably efficient only in the case of solid ^{3}He or in a mixture of liquid ^{4}He with a small admixture of ^{3}He. When solid polarized ^{3}He is rapidly melted in a magnetic field of 5–10 T at a temperature ~ 0.01 K,it is possible to achieve[2,4] $P_n \sim 80\%$, but a polarization at this level is only a transient effect, disappearing rapidly, in 10–300 s, because of relaxation processes.

In their search for an efficient method for producing a steady-state polarized system of ^{3}He, physicists have recently turned to optical methods, which require neither strong magnetic fields nor ultralow temperatures. The optical method of polarizing spin systems, known in the scientific literature as "optical pumping" or "optical orientation," was first proposed back in 1950 by the French scientist Alfred Kastler; for this work he was awarded a Nobel Prize in Physics in 1966. The essence of the method of optical pumping has been discussed repeatedly both in the Soviet literature[5–7] and in foreign journals.[8,9]

In the rf spectroscopy laboratory at the Higher Normal School in Paris, which Kastler directed to the last days of his life, optical pumping was combined with the latest developments in laser technology to produce systems of ^{3}He atoms with an extremely high degree of nuclear polarization in a steady state. A remarkable feature of polarized ^{3}He atoms, which we will denote here by ^{3}He↑, is a uniquely long relaxation time, reaching $\sim 10^5$ s (Ref. 10, for example). This long relaxation time ultimately determines the possible applications of these systems.

Our purpose in the present review (as in other reviews) is to provide a rough guide through the labyrinth of the numerous scientific publications on the topic, so we make no claim that we are offering a comprehensive description of the theoretical models and experimental situations. The structure and content of this review undoubtedly reflect the particular interests of the authors, who are concerned for the

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most part with experimental research. The references at the end of this review include papers published through early 1984, but they of course do not constitute an exhaustive bibliography of all published works.

2. OPTICAL PUMPING IN A SYSTEM OF HELIUM ATOMS

Optical pumping is a method for transferring to atoms an angular momentum carried by the photons of the pump light. The pumping usually involves optical transitions excited between atomic energy levels by polarized resonant light. In the case of ^{3}He atoms, direct optical pumping runs into serious difficulties because the resonant line of He which couples the 1^1S_0 ground state with the nearest (singlet) excited state, 2^1P_1, is in the far-UV region. An "indirect" method for polarizing ^{3}He nuclei was proposed in 1963 by Colegrove *et al.*[11] Their method makes use of two effects: optical pumping in the 2^3S_1 metastable state and spin exchange in collisions between helium atoms in the ground and metastable states.

a) Level diagram and pumping arrangement

The energy levels of the ^{3}He atom between which transitions are used to produce nuclear polarization in the 1^1S_0 ground state are shown by the diagram in Fig. 1. Since the spin of the ^{3}He nucleus is $I = 1/2$, there are two hyperfine sublevels in the 2^3S_1 state, with quantum numbers $F = 1/2$ and $3/2$; in the 2^3P state there are five sublevels: $2^3P_0(F = 1/2)$, $2^3P_1(F = 3/2, 1/2)$ and $2^3P_2(F = 3/2, 5/2)$. A radiative transition between the 1^1S_0 ground state (para-helium) and the 2^3S_1 state (ortho-helium) is forbidden, and the lifetime of ortho-helium, determined by diffusion to the wall, is a few milliseconds. For this reason, the 2^3S_1 state is metastable. Optical transitions can occur between the 2^3S_1 and 2^3P states; they correspond to a resonant line of ortho-helium with a wavelength $\lambda = 1083$ nm. Figure 2 (from Ref. 12) shows the components of the hyperfine structure of this line.

The process by which ^{3}He atoms in the 2^3S_1 state are polarized can be summarized as follows: A glass cell is filled with pure gaseous ^{3}He to a pressure between 0.2 and 5 Torr, and a weak rf glow discharge is excited in it. The purpose of the discharge is to produce metastable ortho-helium atoms.

The relatively long lifetime of the metastable atoms, which is determined primarily by the time required for diffusion to the cell wall, makes it possible to accumulate a significant number of helium atoms in the 2^3S_1 state in the volume; these atoms exist in the presence of a large number of para-helium atoms. As can be seen from Fig. 1, the level diagram of ortho-helium is of such a nature that it is possible to achieve an optical pumping by means of the resonant line at $\lambda = 1083$ nm in this case. For this purpose, a cell holding a mixture of ortho-helium and para-helium atoms, in a weak static magnetic field $\mathbf{B}_0$, which imposes a quantization axis, is illuminated with circularly polarized resonant light in the direction parallel to $\mathbf{B}_0$.

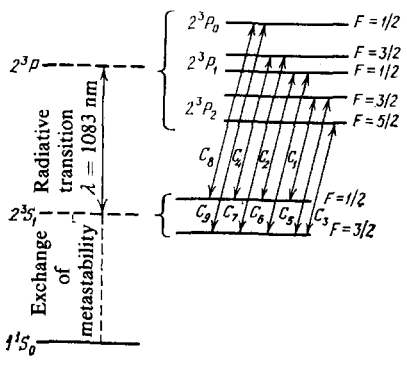

FIG. 1. Energy-level diagram of ^{3}He.

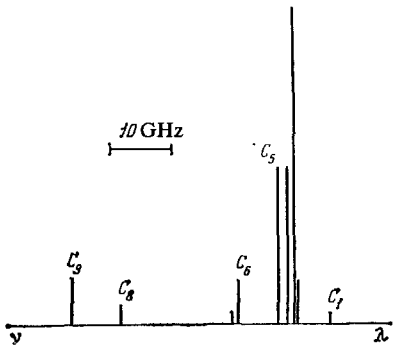

FIG. 2. Hyperfine structure of the emission line $\lambda = 1083$ nm.

To illustrate the optical pumping cycle we consider a pair of levels of ortho-helium, say $2^3S_1(F = 1/2)$ and $2^3P_0(F = 1/2)$, which are coupled by the hyperfine component designated C_8 in Fig. 1. In the magnetic field $\mathbf{B}_0$, both these levels split into two Zeeman sublevels, with quantum numbers $m_F = \pm 1/2$.

For the electric dipole transitions associated with the absorption of a circularly polarized resonant photon (for definiteness, we choose the σ^+ polarization) of the C_8 component there are selection rules which state that an optical transition can occur only from the Zeeman sublevel $2^3S_1(F = 1/2, m_F = -1/2)$ to the sublevel $2^3P_0(F = 1/2, m_F = 1/2)$. Atoms in the 2^3S_1 sublevel $(F = 1/2, m_F = 1/2)$ cannot absorb light in this component of the hyperfine structure at all. Since the inverse transition, $2^3P_0(F = 1/2) \rightarrow 2^2S_1(F = 1/2)$, accompanied by the spontaneous emission of a photon, is allowed to both Zeeman sublevels of the 2^3S_1 state $(F = 1/2)$, ortho-helium atoms accumulate in the 2^3S_1 sublevel $(F = 1/2, m_F = 1/2)$, so that an orientation of the total angular momentum $(\mathbf{F})$ of the metastable atoms arises in the system.

b) Spin exchange between atoms in the ground and metastable states; nuclear polarization of ^{3}He

The weak rf discharge in the cell with ^{3}He and the optical pumping of the ortho-helium atoms by circularly polarized resonance light create a situation in which two spin systems exist simultaneously in the cell: atoms of para-helium (1^1S_0 ground state, with an electron angular momentum $J = 0$ and a nuclear spin $I = 1/2$) and the atoms of ortho-helium, oriented by the optical pumping (2^3S_1 metastable state, with $F = 1/2, 3/2$, where $\mathbf{F} = \mathbf{J} + \mathbf{I}$). The orientation of the total angular momentum in an optical pumping cycle

gives rise[13] to both an electron polarization $\langle \mathbf{J} \rangle$ and a nuclear polarization $\langle \mathbf{I} \rangle$ of a system of atoms. The physical reason for this process is the hyperfine interaction $a\mathbf{I}\cdot\mathbf{J}$, which occurs in both the 2^3P excited state and the 2^3S_1 metastable state. The nuclear polarization $\langle \mathbf{I} \rangle$ which arises in the 2^3S_1 state can be transferred to the 1^1S_0 state of the ^3He atoms in binary collisions

$$^3He\uparrow(2\,^3S_1)+{}^3He\,(1\,^1S_0)\rightarrow {}^3He\,(2\,^3S_1)+{}^3He\uparrow(1\,^1S_0), \tag{2.1}$$

which are called "collisions with an exchange of metastability."[14,15] The interaction between the colliding atoms is electrostatic and does not directly affect the nuclear spin $\mathbf{I}$. If the spin states of the nuclei of the two colliding atoms are identical, such a collision cannot be distinguished from ordinary elastic scattering, since there is no change of any sort in the expectation value of either the electron or nuclear angular momentum in the system. If, on the other hand, the colliding atoms have different nuclear spin states, the result is an exchange of the projections of the nuclear spins.

A detailed theory has been derived[16] for the exchange of metastability in collisions of ^3He atoms, and the effective cross section for this process has been measured: $\sigma_{ex} = (7.6 \pm 0.4)\cdot 10^{-16}\,cm^2$ at $T \sim 300$ K. The corresponding interaction time in the collision is $\tau \sim 10^{-13}$ s, so that we have $\hat{H}\tau \ll 1$, where the Hamiltonian $\hat{H}$ describes the Zeeman and hyperfine interactions of the nuclear spin $\mathbf{I}$. Clearly, under this condition, the nuclear polarization $\langle \mathbf{I} \rangle$ which exists in the 2^3S_1 state is transferred entirely to the 1^1S_0 ground state. This state thus forms a "reservoir" in which the ^3He atoms which are accumulating, with a nuclear polarization, are preserved for a long time.

In most of the experiments which have been carried out so far, the pump light has been provided by a gas-discharge helium lamp with an rf discharge, which emits many lines of the fine and hyperfine structure in the spectrum.

Such light sources have a low power, less than 10 mW in the band of the resonant line at $\lambda = 1083$ nm for the brightest of these sources, so that the nuclear polarization which has been achieved with them has not exceeded $P_n \sim 15$–20% at room temperature and at a ^3He pressure ~ 1 Torr in the cell.[17]

A further increase in P_n required the development of new pump sources, with a high cw output power (hundreds of milliwatts) and a very narrow output spectrum. Progress in laser technology has solved the problem.

c) Production of highly polarized systems of ^3He by laser pumping

It has turned out to be possible to meet these requirements on the source of the pump light by using a laser with an active medium consisting of $(F_2^+)^*$ color centers in a NaF single crystal; this laser was developed a few years ago.[18] Certain modifications in the laser[19,20] made it possible to improve the stability and reduce the spectral width of its output while maintaining a high power level.

Figure 3 is a block diagram of the laser. The $(F_2^+)^*$ color centers in NaF, which have an absorption peak at $\lambda = 870$ nm, are excited by a dye laser, which is in turn

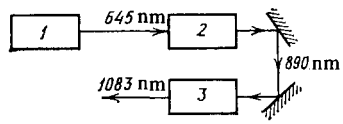

FIG. 3. Block diagram of a laser with a wavelength $\lambda = 1083$ nm. 1—Krypton ion laser, SR 300 K; 2—dye laser, SR 599-01; 3—laser using $(F_2^+)^*$ color centers in NaF.

pumped by the light from a krypton ion laser, emitting in the red lines $\lambda_1 = 645$ nm and $\lambda_2 = 690$ nm. Divalent impurities (Mg^{2+} or Ca^{2+}) in a concentration of 10^{-4}–10^{-3} in the NaF single crystal serve as electron capture centers. The (F_2^+) color centers are produced by bombarding the NaF crystal with an electron beam with an energy of 1.5 MeV and a current density of 4.5 $\mu A/cm^2$ for 40 min at liquid-nitrogen temperature using a Van de Graaf accelerator. The sample is then held at room temperature for 24 h. After this treatment, the single crystals are usually stored at liquid-nitrogen temperature, which is the usual working temperature in the laser method; however, the crystals can also be stored at room temperature for several days without any significant loss of lasing properties.

The ring resonator shown in Fig. 4 is used to operate the $(F_2^+)^*$ laser in a single mode. The NaF single crystal is placed in a vacuum Dewar, cooled with liquid nitrogen over half the distance between spherical mirrors M_2 and M_3, with a radius of curvature $R = 75$ mm.The pump beam from the dye laser ($\lambda = 890$ nm) enters the resonator through beam splitter M_1, and 95% of the intensity of this beam is reflected to the single crystal from mirror M_2. To determine the direction of the ring mode we use a Faraday cell, formed by a glass prism 10 mm long in a magnetic field $B_0 \approx 3.3$ kG. This field rotates the polarization plane by 4°. The rotation is subsequently cancelled by the three mirrors[21] M_4, M_5, M_6. In addition, there are two Fabry-Perot etalons (FP_1 and FP_2) in the resonator; they are required to determine the lasing mode and its scanning. Etalon FP_1 is an air-filled etalon, formed by two prisms with Brewster angles of incidence; one of the prisms can be moved by means of a piezoelectric ceramic, so that we have an electronic self-tuning of the frequency of the mode generated by the laser. Etalon FP_2 is a quartz plate 0.5 mm thick; rotation of this plate along with a rotation of plates P_1 and P_2 makes it possible to continuously tune the frequency.

At a transmittance of 35% of output mirror M_6, this resonator provides an output power of about 300 mW at the

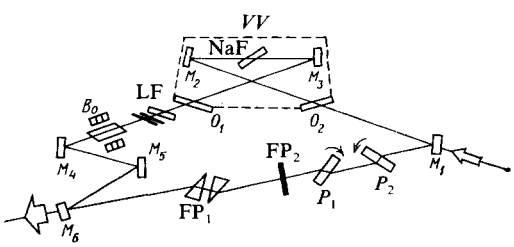

FIG. 4. Optical resonator of the color-center laser. M_1-M_6—Mirrors; FP_1, FP_2—Fabry-Perot etalons; LF—Liot filter; P_1, P_2—rotating prisms; O_1, O_2—optical windows; VV—vacuum volume; NaF—NaF crystal; B_0—Faraday cell.

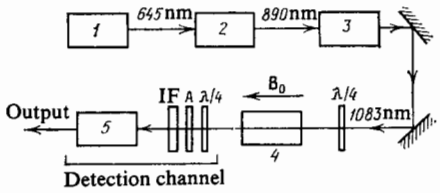

FIG. 5. Block diagram of the apparatus used to polarize ^{3}He nuclei optically. 1—Krypton ion laser; 2—dye laser; 3—color-center laser; 4—cell holding ^{3}He; $\lambda/4$—quarter-wave plate; A—linear analyzer; IF—interference filter for the wavelength 668 nm; 5—photomultiplier.

wavelength $\lambda = 1083$ nm at a pump power of 1.5 W at a wavelength of 890 nm. The output frequency can be tuned continuously over a range of 50 GHz; the instability of this frequency is no greater than 5 MHz.

Figure 1 shows the arrangement of the apparatus for producing nuclear polarization of ^{3}He at room temperature. The pump beam from the laser is expanded and sent through a quarter-wave ($\lambda/4$) plate; it becomes circularly polarized and is sent to a cylindrical cell 5 cm in diameter which is filled with ^{3}He to a pressure ~ 0.3 Torr. This cell is at the center of a Helmholtz coil, which produces a static uniform magnetic field $\mathbf{B}_0 \sim 5$ G, which is directed along the optical axis. The strength of this magnetic field is not important, but it should be as uniform as possible over the cell, since even small gradients of the order of 1 mG/cm sharply reduce the relaxation time of the nuclear magnetization (to ~ 1 min) and lower the maximum degree of polarization $\mathbf{P}_n$.

To detect the signal and to measure the polarization of the ^{3}He nuclei, we use the well-known NMR method, applying an rf magnetic field to the sample.[22] We also use an optical method,[23] which involves measuring the degree of circular polarization of light with a wavelength of 668 nm which is emitted by a discharge in the direction of the field $\mathbf{B}_0$. The components required for this optical method are shown in the measurement channel in Fig. 5: a $\lambda/4$ plate, a linear analyzer A, and an interference filter IF, which singles out the line at 668 nm. These components are positioned in front of a photomultiplier 5. When the $\lambda/4$ plate is rotated at a frequency ω, the photocurrent is proportional to the intensity of the light of alternately left-hand and right-hand circular polarizations, so that the photocurrent becomes modulated at a frequency of 2ω. This modulation makes it possible to use synchronous detection, after which the output signal is directly proportional to the degree of nuclear polarization of ^{3}He.

The results of a theoretical calculation on the kinetics of laser optical polarization in gaseous ^{3}He were reported in Ref. 24. It was assumed there that the frequency of the monochromatic pump light is equal to one of the nine hyperfine components of the line at 1083 nm which are shown in Fig. 2. The interaction of the atoms with the pump photons, the exchange of metastability, and relaxation processes affecting the degree of nuclear polarization were taken into account. Calculations were carried out for the two limiting cases in which there is no disorientation of the atoms in the 2^3P excited state by collisions (very low gas pressures) and in which there is a complete disorientation (high gas pressures). The actual experimental situation of course lies somewhere between these two limiting cases, but a theoretical description of the actual situation runs into serious difficulties. Even in these limiting cases the problem reduces to a numerical integration of a system of coupled nonlinear differential equations.

The calculated results are shown in Fig. 6 as a plot of the steady-state value of P_n versus the power of the laser pump for the case in which the laser frequency coincides with some of the hyperfine components of the helium line. It can be seen from Fig. 6 that the maximum attainable degree of nuclear polarization is significantly affected by both the choice of laser frequency and collisional processes in the 2^3P excited state. The relative importance of the pumping in the different hyperfine components varies in the limiting cases not only in magnitude but also in sign (for the C_1 component, for example). The theoretical conclusions are supported very accurately by data from experiments with single-mode operation of the pump laser.[24]

To maximize P_n, it turns out to be better to use a laser in three-mode, rather than single-mode, operation. In the three-mode case, a very slight change in the construction of the resonator allows lasing on three longitudinal modes, separated from each other by about 150 MHz, near the C_9 component of the hyperfine structure, $2^3S_1(F=3/2) \rightarrow 2^3P_0(F=1/2)$. A mirror beyond the cell reflects the laser beam and makes it possible actually to make use of three more modes, propagating in the opposite direction. This pump arrangement makes it possible to span effectively the Doppler-broadened absorption line of the ^{3}He atoms and to achieve the results shown in Fig. 7 (Ref. 25). The degree of polarization of the ^{3}He nuclei achieved at room temperature in these experiments was $P_n \sim 70\%$ at a ^{3}He pressure ~ 0.3 Torr in the cell and at a laser-beam power of 300 mW.

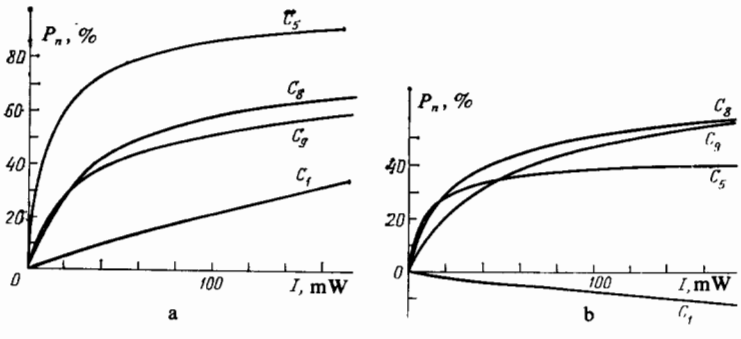

FIG. 6. Theoretical prediction of the degree of polarization of ^{3}He nuclei as a function of the laser beam power in the case of a monochromatic pump at the wavelength of one of several hyperfine components of the ^{3}He line. a—There is no disorientation in the excited state; b—complete disorientation in the excited state.

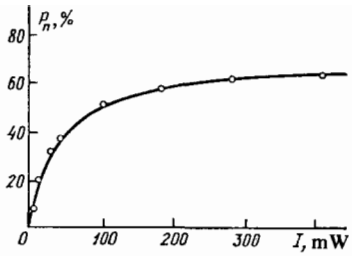

FIG. 7. Degree of polarization of the ^{3}He nuclei as a function of the laser power in the case of pumping at the C_9 hyperfine component of the helium line. Solid line—Theoretical; circles—experimental.

d) Laser pumping at low temperatures

In many physical and technological problems it is necessary to produce a highly polarized system of ^{3}He atoms at low temperatures. The optical polarizaion method is also effective in such cases, at temperatures down to something of the order of 0.01 K. Study of the temperature dependence of the relaxation of optically polarized helium atoms[26–29] shows that the probability for relaxation of the nuclear spins at the cell wall increases rapidly with decreasing temperature. For example, at 77 K the longitudinal relaxation time T_1 is still greater than 1 h, while at 25 K it is $T_1 \sim 1$ s, because the scale time for adsorption of a ^{3}He atom on the cell wall increases rapidly with decreasing temperature. Furthermore, it was found in the papers just cited that the cross section for the exchange of metastability between atoms of ortho-helium and para-helium—this cross section determines the degree of nuclear polarization attainable—decreases by more than two orders of magnitude over the temperature range from 300 K to 4.2 K. These factors make it impossible to achieve values $P_n > 25\%$ at temperatures ~ 4.2 K in glass cells, even when optimum use is made of a laser beam as pump source.[30]

It was found possible to overcome this limitation to a large extent by modifying the cell design and by using two coupled chambers[30,31] (Fig. 8). The upper "warm" chamber, with a volume $V_1 \sim 100$ cm^3, is at room temperature, Θ_1, and communicates through a narrow tube $l \sim 70$ cm long with the lower, "cold," chamber with a volume $V_2 \sim 1$–10 cm^3, which is a cryostat at a temperature $\Theta_2 \sim 4.2$ K. A weak rf discharge is produced in V_1 to produce ortho-helium atoms; for optical pumping of these atoms, a laser beam is directed parallel to the vertically directed static magnetic field $\mathbf{B}_0$. The field $\mathbf{B}_0$ is produced by several (three to five) rings, whose axis is that of the laser beam. The relative positions of the rings and the currents in them are chosen to keep $\mathbf{B}_0$ quite uniform along the tube connecting V_1 and V_2.

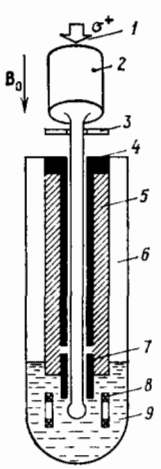

FIG. 8. Cell used for optical polarization of ^{3}He at low temperatures. 1—Pump laser beam; 2—"warm" chamber of cell; 3—mirror; 4—high-temperature heat shield; 5—foam plastic insulation; 6—cryostat; 7—low-temperature heat shield, 8—coils for measuring the NMR signal; 9—liquid helium bath.

The density (n_1) of ^{3}He atoms in chamber V_1 is chosen to maximize the efficiency of the optical pumping: $n_1 \sim 10^{16}$–10^{17} cm^{-3}. Since the diameter of the connecting tube is substantially greater than the mean free path of the ^{3}He atoms, a diffusion equilibrium is established between V_1 and V_2, and the following condition holds:

$$n_1\Theta_1 = n_2\Theta_2. \tag{2.2}$$

The density (n_2) of ^{3}He atoms in V_2 is therefore substantially higher than n_1. At $\Theta_2 = 3$ K, for example, it is possible to achieve $n_2 \sim 10^{18}$–10^{19} cm^{-3}. To increase the nuclear relaxation time T_1 in the cold chamber, a small amount of molecular hydrogen (H_2) is added to this chamber as the cell is being filled. This molecular hydrogen is frozen in the cryostat, and it forms a film of solid hydrogen on the wall of V_2. This film serves as a counter-relaxation coating.

As was shown in Ref. 32, the relaxation time of ^{3}He nuclei on solid hydrogen is several days. Particular attention was paid to the distribution of the gradient along the connecting tube, since this gradient affects the efficiency at which polarized atoms are transported from V_1 to V_2. For this purpose, two copper heat shields are placed in the cryostat. One keeps the temperature of the lower part of the tube near Θ_2, while the other keeps the temperature of the rest of the tube near 100 K. The small distance between these shields minimizes the region with a large temperature gradient, where the polarized ^{3}He atoms could rapidly relax through collisions with the uncoated wall.

Experiments[30] confirm the effectiveness of this cell design. Measurements of the degree of polarization in the warm volume, V_1, were carried out by an optical method, while measurements were carried out in the cold volume, V_2, by an NMR method. It was found that the transfer of the polarization of the ^{3}He nuclei by diffusion from V_1 to V_2 is close to 100% (it is predicted theoretically[30] to be 96%), with the result that a degree of polarization $P_n = 50\%$ is achieved in V_2 at the temperature $\Theta_2 = 4.2$ K, with a density of atoms $n_2 \approx 1.2 \cdot 10^{18}$ cm^{-3}. The relaxation time of the ^{3}He↑ is ~ 6 h. During cw laser pumping of the ^{3}He atoms in the warm chamber, the maximum steady-state value of P_n in the cold volume is reached 1 h after the beginning of the pumping. The cryostat used here made it possible to lower the temperature of the cold volume to $\Theta_2 \sim 1.5$ K over a time interval of 5 min after the maximum polarization of the nuclei was achieved, without any significant decrease in P_n.

These values are not the best possible values, since the gas pressure in the cell could be increased by another order of magnitude without substantially reducing the pump effi-

ciency,[30] and recent experiments[33,34] show that coating the wall with a film of superfluid ^{4}He significantly reduces the relaxation of the ^{3}He nuclei on the wall at temperatures down to something of the order of 0.5 K or lower. Further improvement in the technique for optical polarization of ^{3}He nuclei may result from the use of single-mode cw semiconductor lasers, a gas-compression technique (Ref. 35, for example), or the polarization of a liquid drop through atomic exchange with a polarized gas.[36]

3. USE OF HIGHLY POLARIZED SYSTEMS OF HELIUM ATOMS

The relative simplicity and high efficiency of the optical method for polarizing He in the ground and metastable states are largely responsible for the extensive use of highly polarized helium systems in solving many problems in fundamental and applied physics. The examples which we will discuss below do not exhaust the list of these applications; they simply demonstrate the versatility of the method.

a) Research on exchange and relaxation

As we have already mentioned, a system of ^{3}He↑ atoms has uniquely long relaxation times. A theoretical estimate of the upper limit on the longitudinal relaxation time T_1 under the assumption that the sole relaxation mechanism is the dipole-dipole interaction between ^{3}He nuclei in collisions in the gas phase yields[10] $T_1 \sim 10^6$ s, while the maximum values of T_1 which have been achieved experimentally are $4.5 \cdot 10^5$ s in a cell with a wall coated with solid hydrogen at 4.2 K (Ref. 37) and $7.7 \cdot 10^5$ s in a glass cell with a pressure of 500 Torr at 373 K (Ref. 27). A study of the relaxation of ^{3}He↑ may thus provide a tool for studying extremely subtle and weak interactions in collisions both in the gas phase and with a solid surface; at the same time, such a study is required for solving physical and applied problems in which it is necessary to preserve a nuclear polarization as long as possible.

A detailed examination of all aspects of the relaxation of ^{3}He↑ which have been studied so far would go beyond the scope of this review, so we will simply summarize some of the relaxation mechanisms. We will discuss in more detail the relaxation which occurs at the wall of a cell at a low temperature.

There are at least four mechanisms for the relaxation of ^{3}He↑ which has been polarized in the $1\,^1S_0$ groundstate by optical pumping. The strongest of these mechanisms is "optical" relaxation, which occurs only if atoms of ortho-helium are excited in a cell containing ^{3}He atoms, and they are optically pumped by resonant light. The optical orientation of ortho-helium, followed by an exchange of metastability, described above, is a mechanism which not only serves as a polarizing agent but also reduces the lifetime of the ^{3}He atoms, i.e., causes their relaxation. Optical relaxation has been studied thoroughly (see, e.g., the review by Happer[9]). Its rate is determined by factors such as the intensity and spectrum of the pump light, the parameters of the discharge in the cell containing the mixture of para-helium and ortho-helium, and the effective cross section for the exchange of metastability. Optical relaxation is of course completely ruled out in the absence of a discharge and in the absence of optical excitation.

Another important mechanism is the thermal motion of the ^{3}He↑ atoms in a nonuniform magnetic field. A moving atom "feels" the fluctuating magnetic field, whose Fourier components may cause dipole transitions between magnetic sublevels. This approach has been used to describe the relaxation in a nonuniform field[10,38,39]; the result has been a good agreement between theory and experiment. A more comprehensive theory for the relaxation of ^{3}He↑ in a nonuniform magnetic field was derived in Ref. 40. That theory makes it possible not only to list the restrictions imposed in the earlier studies but also to predict new effects, which stem from the indistinguishability of atoms in highly polarized ^{3}He systems at low temperatures. This type of relaxation occurs in essentially all settings because of the impossibility of producing a perfectly uniform magnetic field. On the other hand, this type of relaxation can be kept at a low level by carefully preparing the experimental apparatus.[37]

A third relaxation mechanism may include various types of disorienting collisions involving momentum transfer in the gas volume. Because of its practical applications, we will expand this case to include both ^{3}He in the $1\,^1S_0$ ground state and metastable ^{3*}He or ^{4*}He in the $2\,^3S_1$ state, where polarization of electron angular momenta can be achieved by optical pumping. When there are impurity atoms or molecules of a different species in the gas volume, binary collisions between oriented helium atoms and the impurity atoms are accompanied by an exchange of spin coordinates (the spin-exchange interaction), and this results in a partial polarization of the impurity atoms. Spin exchange has turned out to be an extremely effective method for polarizing atoms which cannot be optically pumped in a direct way. In particular, this method has been used to polarize He^+ ions,[41] electrons,[42] molecules, and molecular ions.[43] The reader is directed to Ref. 9 for other examples of the use of spin exchange and for a review of the theoretical and experimental results.

The internal energy of the He atom in the $2\,^3S_1$ state is about 20 eV, sufficient to ionize all other atoms except He and Ne. In a collision of an He($2\,^3S_1$) atom with an atom or molecule of an impurity of species X, the following reaction is thus energetically possible:

$$\mathrm{He}\,(2\,^3S_1) + \mathrm{X} \rightarrow \mathrm{He}\,(1\,^1S_0) + \mathrm{X}^+ + e^- + \Delta E. \qquad (3.1)$$

After the collision, the X^+ ion may be in either the ground state or an excited state. An interesting aspect of collisions of this type (Penning ionization) is the possibility of transferring polarization from ortho-helium which has been oriented by optical pumping to either the X^+ ion or the electron produced as a result of the ionization, since the duration of a collision is usually too short ($\sim 10^{-12}$ s) to allow any magnetic interaction between the spins of the particles (the total spin is conserved). A fair number of papers has already been published on interactions of this type, which have rather large effective cross sections in many cases. For example, metal ions,[44] molecular ions,[45] and hydrogen and deuterium atoms[46] have been polarized.

The most complex relaxation mechanism is relaxation

at the wall of a cell containing ^{3}He↑. The van der Waals forces which act between an atom and a wall keep a certain number of the atoms in the immediate vicinity of the wall, with the result that a dynamic equilibrium is set up between adsorbed atoms (τ_a is the mean adsorption time) and free gas atoms. Since there are always microscopic impurities with an electron or a nuclear paramagnetism in a wall, the magnetic interaction, which is effective over a time τ_a, can change the spin orientation of the ^{3}He↑ nucleus; in other words, this magnetic interaction is a cause of relaxation transitions. For this mechanism, the time T_1 would of course have a significant temperature dependence, since this mechanism becomes more effective as τ_a increases, i.e., as the temperature is lowered. Indeed, the measurements carried out in Ref. 47 show that the time T_1 falls off rapidly from 1 h at 77 K to 1 s at 25 K in a glass cell without a coating.

Various types of coatings on the inner surface of the cell wall are used to increase T_1 at low temperatures. These coatings must have a small electric polarizability and small intrinsic magnetic moments. In practice, films of solid hydrogen or of an inert gas are convenient to use at $T < 4.2$ K. These coatings are entities of research interest in their own right, since measurements of the time T_1 can cast light on the interaction of polarized atoms with a surface and on the surface structure.

The relaxation of ^{3}He atoms at a glass wall has been studied[27] over the temperature range 77–350 K, and that on a coated wall has been studied[34,37] at $T < 4.2$ K. The theoretical estimate in Ref. 37 of the probability for longitudinal relaxation at a wall is based on a simple model which has the ^{3}He atoms existing in two phases which are in equilibrium with each other: There are N_V atoms in the gas volume, while N_a atoms are adsorbed on the wall. Their ratio is

$$\frac{N_a}{N_V} = \frac{S\lambda}{V} \exp \frac{\Delta W}{kT}, \tag{3.2}$$

where V and S are the volume and the inner surface area of the cell, $\lambda = h(2\pi mkT)^{-1/2}$ is the thermal de Broglie wavelength, k is the Boltzmann constant, and ΔW is the adsorption energy.

Under the assumption that the adsorption of an atom and its relaxation are independent processes, we can write the relaxation probability $1/T_1$ as the product of the adsorption probability and the probability for relaxation in the adsorbed state, $1/T_a$:

$$\frac{1}{T_1} = \frac{N_a}{N_V} \frac{1}{T_a}. \tag{3.3}$$

In turn, the probability $1/T_a$ is given by[22]

$$\frac{1}{T_a} = \gamma^2 \langle \delta B^2 \rangle \frac{\tau_c}{1+(\omega_0\tau_c)^2}, \tag{3.4}$$

where $\gamma + -2\pi\cdot 3.243$ kHz/G is the gyromagnetic ratio of ^{3}He nuclei, $\omega_0 = -\gamma B_0$ is the frequency of the Larmor precession of nuclei in the magnetic field $B_0 \langle \delta B^2 \rangle^{1/2}$ is the mean square amplitude of the random magnetic field at the wall, and τ_c is the correlation time of the magnetic interaction. We thus have

$$\frac{1}{T_1} = \frac{S\lambda}{V} \gamma^2 \langle \delta B^2 \rangle \frac{\tau_c}{1+(\omega_0\tau_c)^2} \exp \frac{\Delta W}{kT} \approx \frac{S\lambda}{V} \gamma^2 \langle \delta B^2 \rangle \tau_c \exp \frac{\Delta W}{kT} \tag{3.5}$$

under the condition $\omega_0\tau_c \ll 1$.

In expression (3.5), the correlation time τ_c is determined by the physical nature of the interaction which is responsible for the relaxation of the nuclei. This time is short if this interaction fluctuates rapidly over time, while it is quite long if the interaction is quasistatic; in any case, τ_c does not exceed the adsorption time τ_a. Lefevre-Seguin[37] carried out calculations for the particular cases of a strong correlation ($\tau_c = \tau_a$) and a weak correlation ($\tau_c < \tau_a$) of a magnetic perturbation. Different expressions were found for $1/T_1$ in the two cases. For $\tau_c = \tau_a$, the result is

$$\frac{1}{T_1} = \frac{S\lambda}{V} \gamma^2 \langle \delta B^2 \rangle \tau_a^0 \exp \frac{2\Delta W}{kT}, \tag{3.6}$$

where $\tau_a^0 = 4\lambda/\bar{v}\beta$, $\bar{v}$ is the average thermal velocity of the ^{3}He atom at the temperature T, and the coefficient β, which is approximately unity, characterizes the adsorption of an atom on a solid surface.[48] For $\tau_c < \tau_a$, the result is

$$\frac{1}{T_1} = \frac{S\lambda}{V} \gamma^2 \langle \delta B^2 \rangle \tau_c \bar{v} \left(\frac{m^*}{kT}\right)^{1/2} \exp \frac{\Delta W}{kT}, \tag{3.7}$$

where m^* is the effective mass of the adsorbed atoms.

In either case, the temperature dependence of the relaxation probability can thus be described by an exponential factor

$$\frac{1}{T_1} \propto \exp \frac{q\,\Delta W}{kT}, \tag{3.8}$$

where $2 \geqslant q \geqslant 1$.

A systematic experimental study[37] of the temperature dependence of $1/T_1$ at $T < 4.2$ K has confirmed the exponential behavior in cells with walls coated with solid H_2, D_2, or Ne; has made it possible to refine the value of the parameter $q = 2$ in the argument of the exponential function; and has made it possible to determine the adsorption energy in the reactions studied (Fig. 9):

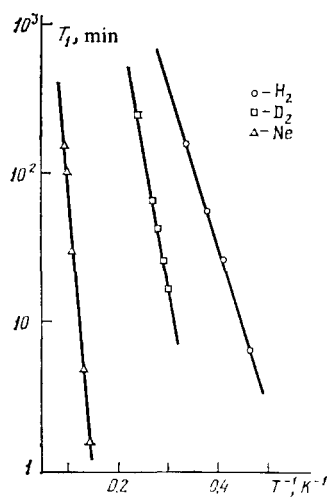

FIG. 9. Temperature dependence of the longitudinal relaxation time of ^{3}He nuclei, T_1, in cells with walls coated with solid hydrogen, deuterium, and neon.

$$\left.\begin{aligned}\Delta W\ (^3\mathrm{He}-\mathrm{H}_2) &= 12\pm3\ \mathrm{K},\\ \Delta W\ (^3\mathrm{He}-\mathrm{D}_2) &= 20\pm3\ \mathrm{K},\\ \Delta W\ (^3\mathrm{He}-\mathrm{Ne}) &= 38\pm5\ \mathrm{K}.\end{aligned}\right\} \tag{3.9}$$

These values, the first found experimentally, agree well with results calculated through a solution of the Schrödinger equation for the ^{3}He atom in the potential of a solid surface containing atoms (or molecules) of hydrogen, deuterium, and neon which are delocalized with respect to the host lattice in accordance with Gaussian law[49]:

$$\left.\begin{aligned}\Delta W_{\mathrm{theo}}\ (^3\mathrm{He}-\mathrm{H}_2) &= 13{,}7\ \mathrm{K},\\ \Delta W_{\mathrm{theo}}\ (^3\mathrm{He}-\mathrm{D}_2) &= 18\ \mathrm{K},\\ \Delta W_{\mathrm{theo}}\ (^3\mathrm{He}-\mathrm{Ne}) &= 37\ \mathrm{K}.\end{aligned}\right\} \tag{3.10}$$

The same measurements yielded an estimate of the mean square value of the magnetic perturbation acting on an He^3 atom during the adsorption time:

$$\langle \delta B^2\rangle^{1/2} \approx 5\ \ \mathrm{G}\ (\text{for } ^3\mathrm{He} \text{ and } \mathrm{H}_2). \tag{3.11}$$

This value is so large that it could be associated only with the field of ferromagnetic microdomains existing in the glass (Pyrex or similar material) with dimensions ~ 100 Å, spaced at distances ~ 3000 Å. These estimates agree with the typical concentration of the impurity Fe_2O_3 in Pyrex ($\sim 0.1\%$).

b) Quantum magnetometers

There are some well-known examples (Ref. 50, for example) of the use of optically polarized ^{3}He and ^{4}He atoms in quantum magnetometry. Optical-pumping magnetometers are used to measure magnetic induction over an extremely broad range, from a few teslas to 10^{-14} T. These instruments are most effective in weak magnetic fields, below 10^{-4} T, and are thus especially pertinent to geophysical and space research. We will skip over the details of the principles underlying the operation of quantum magnetometers with optical pumping—the subject is covered in detail in Refs. 50 and 51—and will discuss here only helium instruments in which the working medium consists of optically polarized ^{4}He and ^{3}He atoms.

1) ^{4}He magnetometers

Since ^{4}He atoms do not have a nuclear spin, and the $1\,^1S_0$ ground state is diamagnetic, magnetometers are constructed by using metastable ortho-helium, which exists in a weak gas discharge. In a magnetic field $\mathbf{B}_0$, the $2\,^3S_1$ state splits into three equally spaced Zeeman sublevels (Fig. 10), whose energy spacing is, in frequency units,

$$f_0\ (\mathrm{Hz}) = (2802350 \pm 30)\ B_0\ (\mathrm{G}). \tag{3.12}$$

Differences are caused in the populations of the magnetic sublevels with $m_F = 0, \pm 1$ by the method of optical pumping with circularly polarized light in the resonant line $\lambda = 1083$ nm, discussed above. The magnetic resonance is excited by imposing an rf magnetic field with a frequency $f_{\mathrm{RF}} = f_0$ on the cell. Changes of two types occur in the system of ortho-helium atoms as a result: The populations of the magnetic sublevels change, and the sublevels acquire a phase coherence. The changes in the populations are seen as

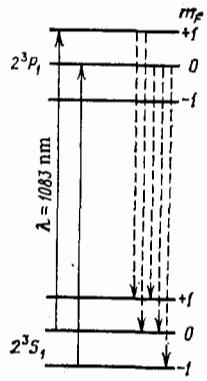

FIG. 10. Energy-level diagram and scheme for pumping ^{4}He atoms by circularly polarized (σ^+) resonance light.

a change in the total absorption of polarized light, while the appearance of a coherence of states generally corresponds to a modulation of the absorption of the light at the field frequency f_{RF}. Optical signals of both types can be used to detect magnetic resonance. Those magnetometers in which a signal proportional to the difference between the populations of magnetic sublevels is detected are by convention called "M_z magnetometers," while instruments which make use of a modulation of the absorption proportional to the phase coherence of the states are called "M_x magnetometers."

Figure 11a is a typical block diagram of an M_z magnetometer. The signal detected by a photodetector is a low-frequency modulation of the total light intensity with the frequency F_{mod}, which is the frequency at which the frequency of the rf oscillator is scanned near $f_{\mathrm{RF}} = f_0$. After amplification and synchronous detection, this signal controls the frequency of an oscillator, automatically tuning it to resonance. The frequency of the oscillator is thus an output parameter, which provides information on the strength of the field being measured.

In an M_x magnetometer (Fig. 11b), the optical signal detected by the photodetector is the modulation of light at the frequency f_0. After amplification and phase correction, the signal is sent to a coil to produce a resonant rf field, thereby providing positive feedback. If the phase is chosen correctly, and if there is sufficient gain in the feedback circuit, continuous self-excited oscillations appear in the system at the frequency determined by the magnetic field B_0.

Helium magnetometers of these two types are the subject of many publications and patents. We might mention the review by Grivet and Malnar[52] who give typical characteristics of M_z magnetometers: a threshold sensitivity $\Delta B_{\min} \sim 10^{-2}$–$10^{-3}$ nT, a signal-to-noise ratio in a 1-Hz band of the order of 10^3–10^4, and a response time (which determines the speed of the instrument) ~ 0.1 s. Some M_z magnetometers are also described in Refs. 53.

Helium M_x magnetometers are described, in particular, in Refs. 54. Their primary advantage is their high speed, which is limited only by the bandwidth of the feedback channel. Furthermore, instruments of this type are structurally simple, and their fabrication is a straighforward process. All helium magnetometers can work over a broad temperature

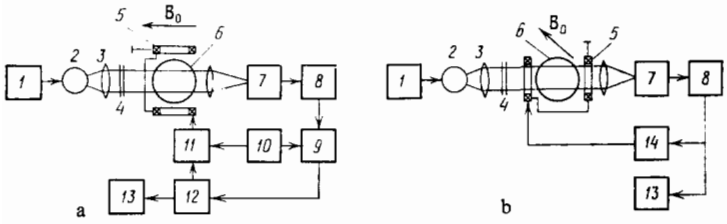

FIG. 11. Block diagram of optical-pumping quantum magnetometers. a: M_z magnetometer. b: M_x magnetometer. 1—Oscillator which excites a discharge in the lamp; 2—spectral lamp; 3—lenses; 4—circular polarizer; 5—rf coils; 6—cell holding the working medium; 7—photodetector; 8—amplifier; 9—synchronous detector; 10—low-frequency oscillator; 11—frequency modulator; 12—high-frequency oscillator; 13—frequency meter; 14—phase-correction circuit.

range (from -60 to $+60$ °C), and they can be rapidly prepared for use after they are turned on, since they do not require the temperature-regulation devices which are necessary for, for example, alkali-vapor quantum magnetometers.

The basic disadvantage of most helium magnetometers is the large systematic error due to optical shifts[55] of the resonance frequency. These shifts depend on the intensity of the pump light, its spectrum, and the orientation of the instrument with respect to the (vector) field being measured. This systematic error is frequently three orders of magnitude greater than the sensitivity.

An effort to eliminate the systematic errors of helium magnetometers while retaining their high sensitivity has recently led to the development of magnetometers with pulsed pumping[56] and alkali-helium magnetometers.[57]

In a helium magnetometer with pulsed pumping, the optical polarization and the measurement of the resonance frequency are carried out at separate times. The pumping is by a periodic train of light pulses, and the magnetic resonance is excited during the dark pause, in which there are no optical shifts of levels. A prototype magnetometer of this design, with a sensitivity of $3\cdot10^{-3}$ nT, has exhibited no optical shifts at the level of 10^{-2} nT.

In the alkali-helium magnetometer, an alkali metal is introduced into the helium-filled cell, and its atoms are subjected to optical polarization. The orientation of metastable ^{4}He atoms in the discharge plasma is caused by Penning collisions with the polarized atoms of the alkali metal, and the resonance of the ortho-helium atoms is detected from the absorption of the pump light by the alkali metal. An M_z magnetometer with a ^{4}He–^{133}Cs mixture has exhibited a sensitivity of 10^{-2} nT with an orientational shift $\sim5\cdot10^{-2}$ nT.

2) ^{3}He magnetometers

The possibility of using optically polarized ^{3}He atoms, which exhibit nuclear paramagnetism in the 1^1S_0 state, is determined by the high efficiency of the mechanism for nuclear polarization through the exchange of metastable states and the exceedingly long relaxation times of ^{3}He nuclei in the absence of a discharge and in the absence of optical excitation. The utilization of these features of ^{3}He↑ in practical magnetometry has taken the path of the development of instruments in which the process of nuclear polarization is separated in time or space from the process of detection of the magnetic-resonance signal. In either approach, optical shifts are competely eliminated, so that magnetometers of this sort are suitable for highly accurate absolute measurements of magnetic fields.

The helium magnetometer which makes use of the free precession of ^{3}He nuclei after the pump light is turned off[58] is similar in signal-detection principle to proton magnetometers (Ref. 50, for example). Because of the high degree of optical polarization of the nuclei, the signal in a ^{3}He magnetometer is usually higher than that in a proton magnetometer, and it decays with a relaxation time three or four orders of magnitude greater than that for the proton signal. It thus becomes possible to carry out prolonged measurements even in a single polarization cycle.

The inconvenience of cyclic operation has been overcome in another version of the instrument, in which self-excited oscillations appear at the precession frquency of ^{3}He nuclei in the field to be measured, B_0 (Maser operation).[59a] Optical shifts are eliminated by using a cell of special design, consisting of two chambers connected by a capillary. The ^{3}He atoms, polarized by the pump light in one chamber, diffuse into the second, where the signal is measured in the absence of a discharge and in the absence of an optical perturbation. The characteristics of the signal in such a cell were also studied in Ref. 59b; operation as a spin generator was achieved in Ref. 59c.

These instruments using ^{3}He have a threshold sensitivity at the level of 10^{-2} nT but are still capable of absolute measurements of the modulus of the magnetic induction. In addition, the small gyromagnetic ratio of ^{3}He nuclei makes systems of this type sensitive to a rotation around the induction vector, so that they are difficult to use on moving platforms.

c) Polarized targets for nuclear physics

Highly polarized ^{3}He systems can be used as targets for studying nuclear reactions or in experiments on the scattering of particles. A target of this type was first used in 1962 to measure the cross section for the scattering of α particles with an energy of 6–7 MeV by polarized helium nuclei.[60] The same target was subsequently used with a deuteron beam in an experimental study of the breaking of P invariance during proton production.

Liquid or solid ^{3}He↑ would naturally be an ideal medium for a nuclear target, but the degree of polarization which has so far been achieved in such targets is less than 1% (Ref. 61). The successful optical polarization of ^{3}He nuclei in a gas at low pressures has made it possible in principle to develop dense polarized targets by compressing a gas polarized before hand by this method.

An extensive program in this direction has recently been carried out at the University of Toronto,[62] where gaseous ^{3}He, polarized by optical pumping, was compressed to 217 Torr by a Toepler compressor. A degree of polarization of 4% has been achieved experimentally at a gas density of $\sim5\cdot10^{18}$ cm^{-3}. The source of the pump light was not a laser

but a discharge helium lamp; this circumstance is one reason for the low nuclear polarization. It is believed that the use of a laser pump and a refinement of the system for purifying and compressing the gas will make it possible to raise P_n substantially.

d) Polarized beams of electrons, ions, and molecules

Many research problems in nuclear physics require beams of polarized particles. Many methods are available for producing such beams, among them the spatial separation of hyperfine states in a strong nonuniform magnetic field,[63] scattering by nuclei,[64] resonance ionization of atoms,[65] and the evaporation of polarized targets.[66] The combination of the technique of optical polarization of ^{3}He atoms with exchange processes during collisions has turned out to be exceedingly successful as a method for producing polarized beams of electrons, ions, and molecules.

The possibility of producing a beam of polarized electrons became obvious immediately after the first experiments on the observation of the polarization of free electrons in a discharge in spin-exchange collisions with optically oriented alkali metal atoms.[67] As was shown later, the polarization of the electrons produced experimentally was retained after their electrostatic extraction from the discharge.[68]

Conservation of spin angular momentum in chemionization reactions involving optically polarized ortho-helium atoms has been used successfully to develop an intense source of polarized electrons[69] suitable for studying some effects which depend on the spin orientation in the scattering of electrons by nuclei, atoms, and molecules at solid and liquid surfaces.

The operation of such a source was described theoretically in Ref. 70; a schematic diagram of the source is shown in Fig. 12. A stream of helium atoms is excited into the 2^3S_1 state as it passes through discharge region 1 in a microwave resonator, and then it enters region 2, where the ortho-helium atoms are optically polarized. During the subsequent motion, the polarized $He(2^3S_1)$ atoms enter region 3, into which CO_2 molecules are simultaneously injected. As the ortho-helium atoms collide with the CO_2 molecules, they

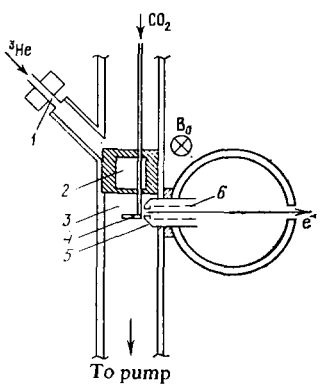

FIG. 12. Schematic diagram of the source of polarized electrons. 1—Region of rf discharge in ^{4}He; 2—region of optical polarization of ortho-helium; 3—region of chemionization; 4—CO_2 ring injector; 5—extractor of polarized electrons; 6—electrostatic lenses. The magnetic field B_0 and the pump beam are both directed perpendicular to the plane of the figure.

ionize the latter in a chemionization process; the electrons which are produced in this reaction and which preserve the spin orientation are extracted by an electrostatic field from the afterglow and are shaped by a system of electron lenses into a collimated beam for further use. Pumping with the light from a helium spectral lamp with rf excitation has produced an electron beam with a current 2 μA and a degree of polarization of $\sim$40% (the electron energy is 100–400 eV).[69] A remarkable feature of this source is the possibility of reversing the electron polarization direction without changing the beam path, by simply rotating the optical polarizer through 90° in the beam of the pump light (by changing the polarization of the light from σ^+ to σ^-). Furthermore, the polarization of the electron beam can be modulated by rotating the polarizer at the appropriate angular velocity.

Further refinement of the source of polarized electrons consisted of the use for optical pumping of a laser with an output in the line at 1083 nm and some structural changes in the afterglow region.[71] As a result, an 80% degree of polarization of the electrons was achieved at a beam current of ~ 1 μA. The modified design of the afterglow tube made it possible to raise the current to 50 μA at a 40% electron polarization. The net result was to achieve a quality factor $P^2I \sim 8\cdot 10^{-6}$ A, which exceeds the characteristics of all known sources of polarized electrons.

Baker *et al.*[72] have reported the development of a source of a beam of polarized ^{3}He ions suitable for use as an injector for an accelerator. The operation of this source is generally similar to that described above, but it uses only a beam of ^{3}He atoms. The metastable ^{3}He atoms ae polarized optically, and the very large cross section ($\sim 10^{-15}$ cm^2) for charge exchange in He^+-$He(1^1S_0)$ collisions makes it possible to achieve a high density of He^+ ions with polarized nuclei. The polarized ions are then extracted from the discharge region by a standard technique, shaped into a beam, and sent to the accelerator. This method has resulted in the production of a beam of He^+ ions with a current up to 4 mA at a degree of nuclear polarization of $\sim$5%. It can be expected that the use of a laser beam as pump will make it possible to substantially increase this degree of nuclear polarization. As in the preceding case, important advantages of this apparatus are its relative simplicity and the possibility of reversing the polarization without changing the particle path.

A similar technique has been used to produce a beam of highly polarized (up to 50%) metastable helium atoms[73] and molecular ions.[45]

e) Other possible applications

Let us take a brief look at some other possible applications of highly polarized ^{3}He systems, as proposed by various investigators. Passell and Schermer[61] have noted that the strong dependence of the cross section for the capture of thermal neutrons by ^{3}He atoms on the orientation of the nuclear spins means that a polarized helium target might serve as a filter for producing polarized neutron beams. This filter might have an advantage over other neutron polarization methods in being applicable over a broad energy range

and over a broad range of the angle of incidence of the neutron flux. Golub[74] has recently shown that the strong dependence of the absorption of ultracold neutrons by ^{3}He nuclei on their spin state might be exploited in an experimental search for an electric dipole moment of the neutron.

An interesting application, although of problematical feasibility, is the idea of polarizing nuclei in a plasma during thermonuclear fusion.[75] It is expected, for example, that the polarization of nuclei of both deuterium and ^{3}He in a reactor with a D–^{3}He mixture would make it possible simultaneously to increase by 50% the cross section for the reaction D–^{3}He and substantially suppress the unwanted D–D reaction. It was originally thought that the relaxation time of nuclear spins in a hot plasma would be too short for this effect to be appreciable; only in recent calculations has it been shown that the nuclear relaxation times may be substantially longer than the scale times of a nuclear fusion reaction.

4. QUANTUM PROPERTIES OF HIGHLY POLARIZED SYSTEMS OF ^{3}He AT LOW TEMPERATURES

a) Theoretical premises

A system of ^{3}He atoms with polarized nuclear spins ($I = 1/2$) has some interesting physical properties at low temperatures, close to the quantum degeneracy temperature. Recent theoretical and experimental studies have made it possible to predict and observe several previously unknown phenomena which depend on the degree of nuclear polarization. Skipping over the details of the theoretical calculations, which can be found in Refs. 76 and 77, we will take a look at the qualitative conclusions of the theories available.

The quantum-mechanical symmetry principle requires that the two-particle wave function $\Psi(\mathbf{r}_1,\mathbf{r}_2)$ of two ^{3}He atoms in the 1^1S_0 state, with parallel nuclear spins, must be antisymmetric under the interchange of these atoms:

$$\Psi_{\uparrow\uparrow}(\mathbf{r}_1, \mathbf{r}_2) = -\Psi_{\uparrow\uparrow}(\mathbf{r}_2, \mathbf{r}_1). \tag{4.1}$$

It follows immediately that we have

$$\Psi_{\uparrow\uparrow}(\mathbf{r}_1 = \mathbf{r}, \mathbf{r}_2 = \mathbf{r}) = 0. \tag{4.2}$$

In other words, two ^{3}He↑ atoms which are polarized in the same direction can never be at the same spatial point; there will always be some minimum distance between the points where the probability for finding the atoms is nonzero. This conclusion is totally independent of the possible existence of any interaction between atoms. The minimum distance to which these atoms can approach each other is some fraction of a de Broglie wavelength, and this minimum distance decreases with increasing kinetic energy of the colliding atoms.

On the contrary, if the nuclear spins of the ^{3}He atoms approaching each other are antiparallel, there is generally nothing to prevent them from occupying the same spatial point, but the repulsive part of the interatomic potential, usually present, will determine the minimum possible distance between them.

In ordinary (unpolarized) ^{3}He, the minimum distance between any pair of atoms is thus determined by the effective range of the repulsive potential, while in 100%-polarized ^{3}He↑ this minimum distance depends on the kinetic energy of the atoms, i.e., on the temperature—increasing with decreasing temperature. In the limit of very low temperatures, this distance in a ^{3}He↑ sample may be totally independent of the interatomic potential.

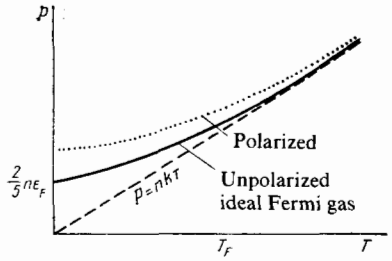

FIG. 13. Diagram illustrating the equations of state of an ideal Fermi gas ($S = 1/2$) in the two cases in which the gas is unpolarized and completely polarized. T_F—Degeneracy temperature; ε_F—Fermi energy.

Two important conclusions can be drawn here. First, the atoms in a ^{3}He↑ system may interact with each other only if they have a sufficiently large kinetic energy, and by lowering the temperature of the polarized atoms one can cause the interaction between these atoms to become infinitely weak (an artificial ideal Fermi gas). Second, for a given density and a given temperature, the kinetic energy of a ^{3}He↑ system is larger than in a system of unpolarized ^{3}He atoms. The latter circumstance is illustrated schematically by Fig. 13, which shows the pressure as a function of the temperature for an ideal Fermi gas, unpolarized or competely polarized, according to the equation of state.

b) Transport processes

What are the consequences of these features of polarized ^{3}He↑ systems? At a very low density, gaseous ^{3}He and ^{3}He↑ are not degenerate and are described by identical equations of state; however, that assertion does not mean that the polarization has no effect on the macroscopic properties of the gas. Obviously, all physical properties for which collisions between atoms are important should be altered when there is a nuclear polarization. For example, the indistinguishability of ^{3}He↑ atoms, which leads to an insensitivity to the interatomic interaction at low temperatures, means that we can expect the mean free path $L(^3\text{He})$ in an unpolarized gas to be shorter than $L(^3\text{He}\uparrow)$ in a polarized gas, and as the temperature is lowered the ratio $L(^3\text{He}\uparrow)/L(^3\text{He})$ should increase (Fig. 14). Since the thermal conductivity K and the viscosity η are proportional to the mean free path, the following inequalities hold:

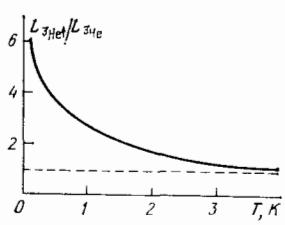

FIG. 14. Temperature dependence of the ratio of the mean free paths of polarized and unpolarized ^{3}He atoms.

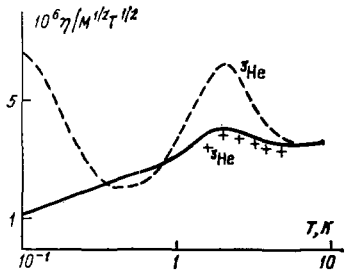

FIG. 15. Temperature dependence of the viscosity η for polarized[76] (dashed line) and unpolarized[78] ^{3}He.

$$K\,(^3\mathrm{He}\uparrow) > K\,(^3\mathrm{He}),$$
$$\eta\,(^3\mathrm{He}\uparrow) > \eta\,(^3\mathrm{He}). \tag{4.3}$$

The dashed line in Fig. 15 is the theoretical behavior of the viscosity as a function of the temperature according to calculations for ^{3}He↑ in Ref. 76. The solid line here shows the theoretical prediction of Ref. 78 for unpolarized ^{3}He, while the points are experimental.[79]

c) Phase diagrams

As the gas density is increased, the difference between ^{3}He and ^{3}He↑ also becomes apparent in the equations of state of the two systems. The first-order correction is usually introduced through the second virial coefficient $B(T)$, which is a measure of the deviation from the behavior which holds for an ideal gas. Figure 16 shows theoretical results[76] on the temperature dependence $B(T)$ for ^{3}He and ^{3}He↑. We see that at any value of T the relation

$$B\,(^3\mathrm{He}\uparrow) > B\,(^3\mathrm{He}) \tag{4.4}$$

holds, and the difference becomes particularly noticeable at $T < 2$ K. Near $T \sim 0.4$ K, polarized ^{3}He↑, in contrast with the unpolarized system, should behave as an ideal gas, while at $T < 0.4$ K the positive value of $B(T)$ for ^{3}He↑ means that the gas pressure is higher at a constant density. In the same temperature region (except at very low T) we have $B(T) < 0$ for ^{3}He, so that the pressure of the unpolarized gas must be lower than that of the polarized gas under otherwise equal conditions.

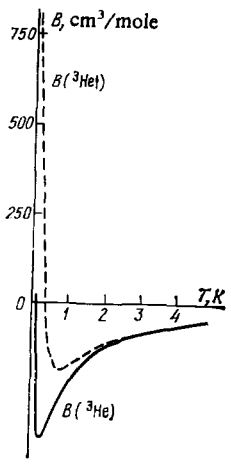

FIG. 16. Second virial coefficient as a function of the temperature for polarized and unpolarized ^{3}He.

Polarized ^{3}He↑ in the liquid phase is predicted to have some interesting features. For example, the existence of nuclear polarization in the liquid state should change the saturation vapor pressure which is at equilibrium with the liquid. Since the binding energy in liquid ^{3}He↑ is smaller than that in liquid ^{3}He, the saturation vapor pressure should be higher in the former case. It can thus be expected that a disruption of the nuclear polarization which exists in the initial state (by an NMR method, for example) in gaseous ^{3}He at a pressure slightly lower than the equilibrium pressure in the liquid-gas system will effectively stimulate liquefaction of the gas.

The significant difference between the physical properties of ^{3}He in the presence of polarized nuclei and the long nuclear relaxation time in both the gas and liquid phases[1)] mean that we can regard the degree of nuclear polarization P_n as a new macroscopic variable which determines the state of a system, along with the pressure P or the temperature T. For example, we can exhibit the qualitative picture of the liquid-gas phase diagram when a partial nuclear polarization is taken into account (Fig. 17). Since P_n is related to the proportion of atoms having the different orientation of the nuclear spin, it is similar to the concentration of one liquid in a mixture of two liquids. We know that the boiling point of a mixture depends on the relative concentration of the components, which changes in the course of boiling. Clearly, the degree of polarization P_n in a gas is higher than that in a liquid phase, so that this unusual phenomenon could in principle be exploited to increase the polarization of gaseous ^{3}He by fractional distillation.

We also know that at temperatures below 0.8 K a liquid mixture of the isotopes ^{3}He and ^{4}He stratifies into two liquid phases. At $T = 0$ K, the upper (and lighter) phase consists of pure ^{3}He, while the lower (and heavier) phase is a roughly 6% solution of ^{3}He in ^{4}He. This lower phase might be thought of as liquid ^{4}He in which a ^{3}He Fermi gas has been dissolved. The properties of polarized ^{3}He↑ discussed above suggest that nuclear polarization would increase the maximum concentration of ^{3}He in ^{4}He. The effect of nuclear polarization on transport phenomena in a liquid ^{3}He–^{4}He mixture has been discussed theoretically.[77,81]

d) Spin waves: experimental results

The propagation of inhomogeneous magnetization oscillations (spin waves) in degenerate liquids is now quite

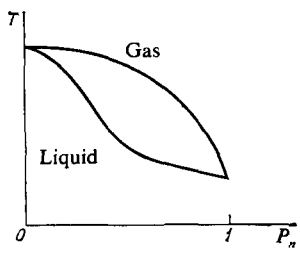

FIG. 17. Qualitative liquid-gas phase diagram for partially polarized ^{3}He at a constant pressure.

well known.[82] The existence of spin waves was predicted as a consequence of the anisotropic part of the molecular field in Landau's theory of a Fermi liquid. It was subsequently shown theoretically [83] and experimentally[84] how spin waves affect spin diffusion in a liquid in spin-echo experiments. Results of a study of spin waves in polarized ^{3}He–^{4}He liquid mixtures by an NMR method[85] and by a spin-echo method[86] have recently been published. Until recently, spin waves have not been observed in polarized low-density gases; it was only just recently that their existence has received unambiguous experimental confirmation.

From the theoretical standpoint, the situation in a gas is different from that in a liquid, since it is not necessary to introduce a phenomenological interaction parameter, and it is possible to calculate[87] all transport phenomena (thermal conductivity, viscosity, spin waves, and so forth) from simply the interatomic potential, for an arbitrary degree of spin polarization ($-1 < M < 1$). It has been predicted that spin waves will substantially change the nature of spin diffusion in a gas even at a relatively high temperature (above 1 K). Introducing the dimensionless coefficient μ, as the ratio of the cross section for the flipping of identical spins to the cross section for collisions between indistinguishable particles,[87] we can write the product μM, which characterizes the relative contributions of the indistinguishable particles to spin diffusion. In a cell in which the lowest-order diffusion mode has a diffusion time τ_D, for example, the frequency of a transverse spin wave in a coordinate system rotating at the Larmor frequency is given by

$$\delta\omega = \frac{\mu M}{\tau_D}. \tag{4.5}$$

Measurements of the coefficient μ make it possible to test the theoretical assumptions and to obtain information on the interaction potential.

The first successful experiment on the observation of spin waves in gaseous ^{3}He↑ in the temperature interval 2–6 K was recently carried out in a laboratory at the Ecole Normale Superieure in Paris.[88] The technique of laser optical polarization of ^{3}He nuclei at low temperatures, described above, was used to produce a polarized system of helium atoms. Spin waves were excited in the sample by imposing a magnetic-field gradient δB_0. The effect of the spin waves was seen, in agreement with the theory of Ref. 40, as changes in the decay parameters of the transverse-magnetization signal M_+, specifically, changes in the transverse relaxation time T_2 and the shift of the precession frequency, $\Delta\omega$:

$$\frac{1}{T_2} \approx a\,|\,\delta B_0\,|^2\tau_D,$$
$$\Delta\omega \approx a\,|\,\delta B_0\,|^2\tau_D\cdot\mu M. \tag{4.6}$$

The quantity μ can be related directly to the measured quantities:

$$\Delta\omega T_2 = \mu M. \tag{4.7}$$

Because of the high degree of polarization of the ^{3}He ($\sim 30\%$) and the high signal-to-noise ratio, it was possible to detect exceedingly small relative changes in the precession frequency of the ^{3}He nuclei (about 10^{-3} Hz at an average frequency of $1.2\cdot10^4$Hz). A method of extrapolating the

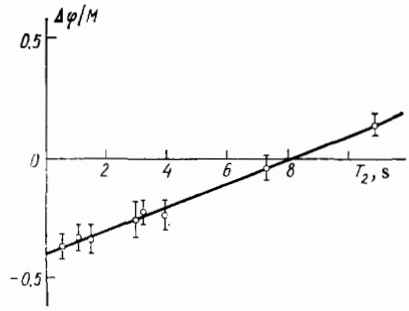

FIG. 18. Phase shift of the free-precession signal as a function of the transverse relaxation time T_2 in a nonuniform magnetic field.[88]

measured values of $\Delta\varphi/M$, where $\Delta\varphi$ is the phase shift, to a zero value of T_2 (i.e., to the limit $\delta B_0 \to \infty$) was used to separate the frequency shifts due to spin waves and the shifts caused by other factors (the response of the receiving coil, spin diffusion in the connecting tube, the effect of the superconducting screen, etc.). Figure 18 shows some of the results obtained by this method, while Fig. 19 shows the temperature dependence of the coefficient μ. The solid line is the theoretical curve of Ref. 87, while the points are the experimental values.

The methods which have been used to study the propagation of spin waves in nondegenerate polarized gases have also proved effective for describing condensed systems (a solution of ^{3}He↑ in superfluid ^{4}He).[89] Calculations of the spectrum of spatially homogeneous magnetization oscillations in a two-component paramagnetic gas in an external field agree well with experimental data on both[88] ^{3}He↑ and a dilute solution[86] of ^{3}He↑ in ^{4}He. Similar effects were recently observed[90] in polarized hydrogen, H↑, demonstrating that the properties of spin-polarized quantum systems are quite general.

Collective effects in such systems are extremely interesting for fundamental and applied physics. For example, Bashkin[91] has shown that magnons would have a dramatic effect on the thermodynamics of liquid ^{3}He↑, and the competition of the magnon and fermion contributions to the free energy would lead to a spontaneous ferromagnetic ordering in liquid ^{3}He↑ at nonzero temperatures. This new and rapidly growing branch of the physics of low temperatures has already become well established as a direction in current research.

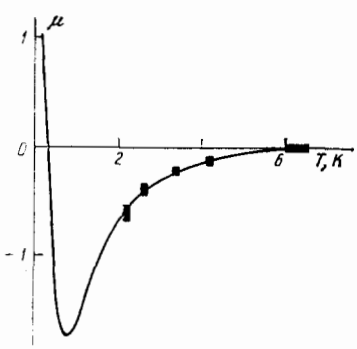

FIG. 19. Comparison of the experimental values of the coefficient μ with the theoretical prediction of Ref. 87 (solid line) at various temperatures.

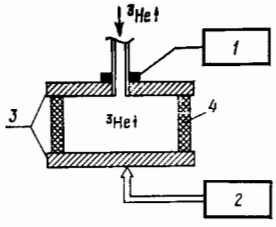

FIG. 20. Schematic diagram of a planned experiment to study the thermal conductivity of polarized ^{3}He. 1—Heater; 2—temperature-sensitive element; 3—copper plates; 4—heat insulator.

e) Planned experiments

As we have already mentioned, the polarization of nuclear spins in ^{3}He should strongly affect transport phenomena in the gaseous and liquid states. Although these conclusions have yet to receive experimental confirmation, the high degree of polarization achieved through the use of laser optical pumping and the long relaxation times of ^{3}He↑ nuclei which have already been achieved raise the hope that experiments of this type will be successful. Preparatory work is already being carried out on a study of the thermal conductivity of ^{3}He↑ at low temperatures. The experimental arrangement is shown schematically in Fig. 20. This experiment will require the development of a special cell which will be filled (by the technique described above) with a polarized ^{3}He↑ gas of sufficient density at a temperature $T < 4.2$ K. The preliminary results show that success is possible. A next step in this direction will be the study of the viscosity and other transport properties of highly polarized quantum systems.

5. CONCLUSION

The high efficiency of optical methods for polarizing helium atoms make them useful for an extremely wide variety of projects in fundamental and applied research. The future development of these methods will proceed hand in hand with progress in laser technology, in particular, the development of new semiconductor lasers.

In this review we have attempted to demonstrate the actual capabilities of optical polarization methods. These capabilities are still far from being exhausted. The combination of these methods with advanced experimental technology will undoubtedly lead to many new and interesting results.

[1)]The relaxation time of ^{3}He nuclei dissolved in liquid ^{4}He was measured in Ref. 80 and found to be 128 ± 8 h in a cell with a wall coated with solid hydrogen.

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