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QUANTUM PROPERTIES OF SPIN POLARIZED ${}^{3}H_{e}$ (${}^{3}H_{e^{\dagger}}$)

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Résumé.- Les effets d'une forte polarisation nucléaire dans un ensemble d'atomes de ³He sont discutés théoriquement. L'indiscernabilité des atomes entraîne que cette polarisation nucléaire augmente leur énergie cinétique et réduit le rôle de leurs interactions. En conséquence, les propriétés macroscopiques de ³He gazeux, liquide ou solide peuvent être changées de façon appréciable. Plusieurs exemples sont succinctement discutés : modifications des propriétés de transport de ³He gazeux à quelques degrés Kelvin, du diagramme de phases d'équilibre liquide-gaz ou liquidesolide, etc.

Abstract.- The effect of a high polarization of the nuclear spins in an ensemble of ³He atoms are theoretically investigated. One can see from the Pauli antisymmetrization principle that a non-zero nuclear polarization results in an increase of the kinetic energy of the atoms and in a decrease of the effectiveness of their interactions. As a consequence, the macroscopic properties of gaseous, liquid or solid ³He at low temperatures may be significantly altered. Several examples of these changes are briefly discussed in this article : modifications of the transport properties of gaseous ³He at a few degree Kelvin, of the liquid-vapour and liquid-solid phase diagram, etc.

1. Introduction.- Let us consider two ³He atoms, both in the electronic ground state. If we assume that their nuclear spins (I = 1/2) are parallel, the symmetrization principle of quantum mechanics requires that the two atoms wave function, $\psi(\vec{r}_1, \vec{r}_2)$, be antisymmetrical in the exchange of the two atoms :

$$\psi_{\uparrow\uparrow}(\vec{r}_1, \vec{r}_2) = -\psi_{\uparrow\uparrow}(\vec{r}_2, \vec{r}_1)$$
(1)

Since :

 $\psi_{++}(\vec{r}_1 = \vec{r}, \vec{r}_2 = \vec{r}) = 0$ (2)

the atoms can never be found exactly at the same point of space. As a consequence, there is some minimum distance between two points where the two atoms can be observed with a non-negligible probability. This result is independent of the existence of any interaction between the atoms ; we shall see below that the minimum distance in question depends on the relative kinetic energy of the atoms. It is easy to understand that this phenomenon occurs only because the two atoms have parallel spins : if they were antiparallel (nuclear spins in orthogonal quantum states), there would be no reason in general why the atoms should not occupy the same point of space.

What happens now if we try to force two ³He atoms with parallel spins to come very close ? We can for example produce a headon collision between two atoms and expect that, during the collision time, their distance will become very small. Figure 1 shows pictures describing what actually happens, for non-interacting one dimension gaussian wave packets. We see that, during the collision, that is when the two wave packets overlap, guantum interference effects occur which ensure that the atoms never come very close to each other. The minimum distance of approach is some fraction of the De Broglie wavelength (which is the shortest length we have introduced in the problem). If we want to reduce the minimum distance between the atoms, we necessarily have to reduce their De Broglie wavelength, which increases their kinetic energy.

If now the nuclear spins are antiparallel, instead of parallel, figure 1.a has to be replaced by figure 2 : no interference effects occur so that the atoms can be found very close from each other during the collision (whatever their kinetic energy)^{*} . In practice, the repulsive part of the interatomic potential will of course put a lower limit on their relative distance, but this is a different physical phenomenon.

The aim of the present article is to study the differences between the properties of ordinary ³He, with nuclear spins pointing randomly in all directions, and spin polarized ${}^{3}\text{He}$ (or ${}^{3}\text{He}\uparrow$), i.e. an ensemble of atoms with all nuclear spins parallel to each other. Of course, macroscopic samples of ³He or ³He⁺ contain a very high number of atoms, and the preceding discussion, in terms of two atoms only, is not sufficient. Nevertheless, the physical ideas we have obtained remain valid : in ordinary ³He, the minimum distance at which any pair of atoms can be found is simply determined by the range of the repulsive potential. On the other hand, in ${}^{3}\text{He}^{\dagger}$, this distance also depends on the kinetic energy of the atoms and increases when this energy decreases ; in fact, at very low energies, the minimum distance between atoms becomes completely independent of the interatomic potential. We can obtain the following general conclusions from the preceding discussion : (i) The polarization of the nuclear spins reduces the effects of the atomic interactions.

In ${}^{3}\!He+$, the atoms can only interact as far as their kinetic energy is high enough. For example, if this kinetic energy becomes very low, the De Broglie wavelength will exceed the interatomic potential range a_{o} , so that the effect of the potential will be masked (the atoms are never close enough to interact). The minimum distance of approach which can be seen in figure 1.e is nothing but the size of the so called "exchange hole". Therefore, it is equivalent to say that, at very low kinetic energies, the exchange hole strongly reduces the effective interactions between the atoms.

As an example, let us consider a low density gas of ${}^{3}\text{He}*$, and compare it to a gas



Fig. 1.- Wave packets of two colliding ³He atoms with parallel nuclear spins. Several simplifying assumptions have been made in the computation of these figures : the wave packets are Gaussian, they have no y-z dependence (one dimension problem), and the effect of the interactomic potential is ignored. The curves show the probability density associated to the "relative particle", with position $x = x_1 - x_2$ (x_1 and x_2 give the positions of the two atoms).

Figure a shows the wave packets before colli sion ; λ_{DB} is the De Broglie wavelength of the atoms. When the two wave packets come closer and closer, interference effects become gradually more and more important (fig. b to e) ; they ensure that the probability density at x = 0 always remains zero : both atoms can never be found at the same point of space. The region around x = 0 where the probability is negligible is nothing but the "exchange hole", which has size comparable to the De Broglie wavelength.

^{*} A third possibility occurs when the nuclear spins are in the singlet state, which is a coherent superposition of states where each atom has a well-defined spin direction. Interference effects then occur again, but with a different phase from fig. 1-e : the interference is now constructive at x=0 (as for spinless bosons).

of ordinary ³He at the same density and temperature T. In a dilute system, the kinetic energy of the atoms depends linearly on T (their De Broglie wavelength is proportional to $T^{-1/2}$). Therefore, at very low temperatures, the nuclear polarization can be used to render the atomic interactions completely negligible ; in this way, an "artificial ideal gas" is obtained.

Fig. 2 : This figure is similar to Fig.1-e but it has been assumed that the atoms have antiparallel nuclear spins (for example, the spin of the atom coming from the right points upwards, the spin of the other downwards). No interference effect then occurs and the atoms can be found at the same point of space. A short range interatomic potential would be more effective in this situation than in the situation of fig. 1-e.

(ii) At given density, the nuclear polarization increases the kinetic energy of the system.

We now want to discuss kinetic energy effects which are increasing functions of the atomic density. Let us qualitatively discuss what happens when we increase the density of a system made of a high number of atoms. Of course, we can always squeeze two (or more) atoms in the same small region of space, but figure 1 shows us what then happens : this operation reduces the period of oscillation of the interference term, and we know that fast oscillations of the wave function always imply a high kinetic energy for the particles. Clearly, this effect occurs in both ³He and $^{3}\text{He}^{4}$, since in ^{3}He the spins of any given pair of atoms may also be parallel. The point is that the effect is more pronounced in ${}^{3}\text{He}^{+}$ because all pairs of atoms have parallel spins. This phenomenon can be

illustrated by a very simple case : the ideal gas (no interactions). Polarizing

all nuclear spins will divide by two the one atom density of states, so that the Fermi level E_F will be raised ; for example, at a given density and zero temperature, E_F will be multiplied by a factor $2^{2/3}$. As a consequence, the equation of state of the gas will be modified, as shown in figure 3: at given T and atomic density, the gas pressure is increased by the nuclear polarization.





Fig. 3 : Diagrams showing the equation of state an unpolarized ideal gas, compared to a fully polarized gas at the same density. The role of the Pauli exclusion principle is enhanced by the nuclear polarization, which results in a higher pressure.

The rest of this article is a discussion of the consequences of these two physical effects on the properties of real ³He+, as compared to ordinary ³He. We shall first study the dilute phases, that is gaseous $^{3}\text{He}^{+}$, and discuss the modifications of the gas properties induced by a 100% nuclear polarization. Then we shall study liquid and solid ³Het, which raise more delicate, but interesting, questions. We shall only discuss here the main physical effects, without any detailed calculations ; more details can be found in ref. /1/. The reader is also referred to ref. /2/, where the thermodynamical aspects of these problems are discussed and more emphasis is given on the properties of dense phases. 2. Properties of gaseous ³Het. - 2.1. Very dilute gas. - A very dilute sample of ³He, or³He⁺, is non-degenerate , so that at the low density limit, gaseous $^{3}\mathrm{He}$ and $^{3}\mathrm{He}^{4}$ have the same equations of state. This does not mean that the nuclear polarization of the atoms has no macroscopic effect on the gas, and we shall see several examples where they are indeed important. It is immediately obvious from figure 1-e and 2 that all physical properties of the gas which depend strongly on collision phenomena may change under the effect of nuclear polarization.

For example, one can think of a scattering experiment (neutron scattering for example) where the properties of very close pairs of atoms (or ³He-³He transient molecules formed during collisions) are observed ; it is clear that the oscillations of figure 1-e will affect the value of the two body density at short interatomic distances and therefore change the properties of the light scattered by close atomic pairs (in ³He⁺, the interatomic distance in the transient molecules will be larger than in ³He). In this section, we shall rather focus the interest on another kind of macroscopic properties of a ³He gas which strongly depends on collisional effects : the transport properties, heat conduction and viscosity (which are purely non-equilibrium properties, in opposition to the two body density). The simplest approach to study these transport properties consists in using the so called mean free path theory. We shall use the letter & for the mean free path in ${}^{3}\text{He}$, and $l \uparrow$ for the same quantity in ³Het.

According to the discussion given above, the interactions between the atoms are masked in ³He+ by particle indistinguishability effects.

Therefore, we expect that :

lt > l

The lower the temperature, the longer the De Broglie wavelength of the atoms, and the higher the value of the ratio l^{\dagger}/l . Since the heat conduction coefficient K and the viscosity coefficient μ are both proportional to the mean free path in the gas, we expect that :

μ† > μ

A precise calculation of these quantities

is given in ref. /1/, adapting to ³He⁺ the calculations of Munn et al./3/. Figure 4 shows the results obtained for the coefficients K and μ ; the qualitative arguments given above are indeed vindicatived and, at temperature T \leq 1 K, significant differences between ³He and ³He⁺ are predicted.



Fig. 4 : Variations of the gas viscosity μ (or the heat conduction coefficient K) of normal ³He and ³He⁺, as a function of the temperature T. The quantity actually shown is μ/\sqrt{MT} where M is the molar of ³He, since this number is temperature independent for a classical hard sphere gas. The full line gives the theoretical predictions of Monchick et al./3/, the crosses the experimental results of Becker et al./4/, both for normal gaseous ³He. The broken line gives the results of our calculations for ³He⁺. Two new effects appear at a 100% nuclear polarization :

(i) in the region 2 < T < 4 K, a strong quantum oscillation (quantum diffraction effect), which was hardly visible in the case of ordinary ³He.

(ii) a low temperature divergence, arising from the absence of any s-wave scattering in $^3\mathrm{He}^{\star}.$

Table I

т	300 K	4.2 K	2 К	0.1 K
L(³ He)	2.4 cm	1.5 cm	1.7 cm	0.5 cm
L(³ He↑)	2.4 cm	1.8 cm	2.6 cm	3 cm

Table I gives the values of l and $l\uparrow$ at various temperatures. We see that, at room temperature, the mean free path of ³He \uparrow and ³He are practically the same, but that they become significantly different at low temperatures.

<u>2.2. Denser gases</u>.- In denser gases, the differences between ³Het and ³He also appear in the equation of state of both systems. The first order correction in density of a gas is usually expressed in terms of a virial coefficient B(T), which characterizes the deviation from the ideal classical gas law. A positive value of B(T)implies a higher value for the pressure (at given density and temperature) and is a consequence of either the repulsion of the atoms, or of their Fermi statistics. A negative value of B(T) in a fermion system is due to the attractive part of the interatomic potential /5/.

Figure 5 shows the calculated values of B(T)for ³He and ³He⁺ as a function of the temperature T [these numerical values are obtained by a generalization of the results



Fig. 5 : Variations of the second virial coefficients B(T) as a function of temperature. The full line gives B(T) for ordinary ³He, the broken line for ³He4 . When T \simeq 0.4 K, ³He4 is very similar to a classical ideal gas, but not ³He. At very low temperatures, the Fermi statistics effects dominate and both coefficients B(T) become larger and larger (but the virial development can no longer be limited to its first term).

of Boyd et al. /6/. We see that, as expected, B(³He⁺) is always greater than B(³He); as a matter of fact, these coefficients are

significantly different at any temperature $T \leq 2$ K. A more detailed discussion of these differences is given in ref. /l/, in terms of the effect of the nuclear polarization on the kinetic energy of the atoms (ideal gas term) and on the atomic interactions.

3. <u>Properties of liquid ³Het</u> -- Normal ³He forms a very weakly bound liquid : the binding energy per atom is the sum of a kinetic energy $E_c \approx 10$ K and a potential energy $E_p \approx -12$ K, and therefore results from a delicate balance between larger energies. Since E_c will be higher in liquid ³Het than in ordinary ³He, one may even ask if liquid ³Het will remain stable when fully polarized.

Any liquid can be considered as an enormous cluster of many atoms. It is therefore interesting to have an idea of the minimum number n⁺ of ³He⁺ atoms which form a bound state (if n⁺ = ∞ , liquid ³He⁺ does not exist). A discussion of this question is given by T.K. Lim et al. in the same journal issue /7/ : at least n=12 unpolarized ³He atoms are probably needed to form a cluster, and n⁺ is even larger.So, it is not easy to bind together several ³He⁺ atoms.

In ref. /1/, we give a simple estimation of the difference between the binding energy of the two phases (this difference is extrapolated from its low nuclear polarization value, which is known from the magnetic susceptibility of the liquid). If this result is correct, the energy variation is only one tenth of the total binding energy, which means that the nuclear polarization effects are ten times too small to prevent the formation of a liquid.

Much more elaborate methods to discuss this problem have been used by J.W. Clark et al., M. Ristig and P. Lam, M. Miller and R. Guyer, and one of us in collaboration with D. Levesque. They are discussed in the corresponding articles in this journal issue /8/ /9/, /10/,/11/. From these studies, a better knowledge of both liquid ³He and ³He⁴ should result.

Figure 6 shows how the nuclear polarization of the ³He liquid should change it saturating vapour pressure : liquid ³He⁺ being less bound than liquid ordinary ³He, its vapour pressure should be higher. Therefore, starting with a ³He+ gas at a very slightly lower pressure than the liquid-gas equilibrium pressure, it should be possible to trigger a liquefaction phenomenon (formation of droplets) by destroying the nuclear polarization (e.g. by N.M.R. techniques).



Fig. 6 : Approximate variations, as a function of temperature, of the ratio between saturated vapour pressures of ${}^{3}\text{He}^{4}$ and ${}^{3}\text{He}$.

The liquid-gas equilibrium also shows interesting properties when the nuclear polarization X is only partial (X = 50% for example). Since X is simply related to the proportion of spin up and spin down atoms, X is somewhat analogous to the concentration of a substance A in a mixture of A and B. We know that mixtures of two liquids do not boil at a fixed temperature, and that the concentrations in both phases change during the ebullition. Figure 7 gives a sketch of the liquid-gas equilibrium diagram of partially polarized ³He. When the two phases coexist, the polarization X is higher in the gas than in the liquid phase, where a strong nuclear polarization implies a larger energy increase than in the gas. This unusual phenomenon could, at least in principle, be exploited to increase the polarization of a partially polarized ³He sample by the methods of fractional distillation.

Until now, we have discussed only the equilibrium properties of liquid ³He+. It is nevertheless clear that, like in the gas phase, the transport properties (sound velocity, etc..) should also be affected by a nuclear polarization. Liquid ³He-⁴He solutions also provide us with interesting physical systems where the effects of a nuclear polarization should be significant. Very long nuclear relaxation times have been obtained in this system by M. Taber et al. /12/. We know that two phases can coexist at low temperatures : a pure ³He phase and a mixed ³He-⁴He superfluid phase with a fixed ³He concentration (6% at zero temperature). A simple model



Fig. 7 : A sketch of the phase diagram of the liquidvapour equilibrium for partially polarized 3 He, at constant pressure. Ordinary 3 He corresponds to X=0, fully polarized 3 He to X=1. When the liquid and vapour phases are in equilibrium, the polarization X is higher in the latter phase. This is because a given polarization costs more energy in a dense phase than in a dilute, non degenerate, phase.

assimilates the latter to a gaseous ³He phase and allow us to extrapolate the prediction given above concerning the liquidvapour equilibrium : the nuclear polarization should increase the maximum concentration of ³He in ⁴He. The existence of a tricritical point also raises interesting question : how will this point move when the nuclear polarization is 100% ? It is also clear that, when the nuclear polarization X is only partial, it should take on different values in the different phases, so that interesting phase diagrams as a function of X should occur. The effects of a nuclear polarization on the transport properties of liquid ³He-⁴He mixtures are also spectacular, as shown by E.P. Bashkin and A.E. Meyerovich /13/. To conclude this section, we can remark

that a full nuclear polarization in a ³He system considerably simplifies the problem of finding the energy levels and the associated orbital wave functions. In ordinary

$$\psi > = |\psi_{\text{orb}} > \otimes |\psi_{\text{spin}} >$$

where $|\psi_{orb}\rangle$ and $|\psi_{spin}\rangle$ are kets describing respectively the orbital and spin variables of the system. This impossibility arises from the complicated structure of the antisymmetrical state space, which is not simply a tensor product of an orbital state and a spin state space. Of course, we can always write

$$|\psi\rangle = \sum_{n} C_{n} |\psi_{orb}^{(n)}\rangle \otimes |\psi_{spin}^{(n)}\rangle$$

but, then, an eigenstate of the system is associated to several orbital states $|\psi_{orb}^{(n)}\rangle$. The situation is therefore much more complicated than for an ensemble of ⁴He atoms, which can be described by one ket $|\psi_{orb}\rangle$ (i.e. one wave function). In fully polarized ³He⁺, this problem disappears and the state vector always factorizes :

$$|\psi\rangle = |\psi_{orb}\rangle |S = N/2, M_{g} = N/2 \rangle$$

where $|\psi_{orb}
angle$ is a state vector belonging to the space of orbital variables and |S = N/2, $M_{s} = N/2$ the fully polarized spin state (the quantization axis is chosen parallel to the spin). The ket $|\psi_{orb}\rangle$ is equivalent to a wave function depending on 3N variables, the coordinates of the particles. The mathematical problem of finding the ground state level and the first excitations of the system is then similar to the same problem of a system of spinless bosons (⁴He atoms), the only difference being that the wave function is now antisymmetrical by exchange, instead of symmetrical (*). This should strongly reduce the spectrum of elementary excitations in liquid ³He. Superfluidity phenomena should then be markedly different in ³He[†] than ³He, and appear at significantly higher temperatures in the first case.

Even in the absence of superfluidity, one may consider both system as two distinct Fermi liquids with their own characteristics (specific heat, sound velocity, etc...) Even if the macroscopic properties of fully polarized ³He do not differ dramatically from those of unpolarized ³He, ³Het provides us with a "new" physical system which may be easier to understand from a microscopic point of view than an ordinary ³He. For example, variational calculations should be more accurate and give a better description of reality in ³He⁺ than in ³He : this is again because the full antisymmetrization puts a very strong constraint on the orbital wave function and leaves much less freedom for the choice of possible wave functions.



Fig. 8 : The changes in the phase diagram of ${}^{3}\text{He}$ created by a 100% nuclear polarization are schematically shown in this figure. Full lines : ${}^{3}\text{He}$; broken lines : ${}^{3}\text{He}^{\dagger}$. The nuclear polarization reduces the domain of existence of the liquid phase.

4. Other phases : solid, adsorbed films, etc..- In solid ³He, the atoms are relatively well localized on a crystal lattice, and it is well known that the exchange effects

^(*) In terms of group theory, for normal 3 He the energy levels correspond to orbital wave functions which span multidimensional representations of the permutations of the particles. For 3 He⁺, as for 4 He, only one dimension representations are useful.

are weak compared to the same effects in the liquid. We therefore expect that the consequences of the nuclear polarization will be weaker in the solid than in the liquid ; in other words, the kinetic energy per atom is relatively high in the solid anyway, and the nodes of the wave function of ³He⁺ do not imply very different values for the energy of the system. The situation is then analogous to the gasliquid equilibrium : in ${}^{3}\text{He}^{4}$, the domain of existence of the solid exceeds the domain for normal ³He. One can also predict the disappearance of the Pomeranchuk effect, which arises from a higher nuclear spin entropy in the solid ³He than in liquid ³He. Obviously, this effect does not occur in ³Het, where there is no nuclear spin entropy anyway. Another extremely interesting feature is the possibility, suggested by B. Castaing and P. Nozières /2/ that solid ³He⁺ could be a vacancy solid as introduced by Andreev /14/.

There are other situations where the study of the properties of ³He⁴ could be interestinf. We have already mentioned ³He⁴-⁴He mixtures, and one can envisage other systems, like two dimensional ³He⁴ (i.e. ³He⁴ adsorbed on a solid surface like graphite or on a liquid ⁴He surface), etc.

5. <u>Conclusion</u>.- Spin polarized ³He provides us with an attractive physical system which exhibits several interesting quantum features. Of course, the equilibrium states of ³Het are in fact only metastable states, but, as far as the nuclear relaxation time T_1 is long enough, the nuclear polarization X can be considered as a new macroscopic variable, like the pressure P or the temperature T. Phase diagrams including the variable X exhibit interesting phase changes and, since these diagrams are not precisely known at the present time, they seem to be worth studying.

The main practical problems in observing ³Het experimentally are to produce this system and to keep it polarized (long relaxation times). Several polarization methods have been proposed : some use the so called "brute force technique", like the fast melting of the solid proposed by Castaing and Nozières /2/, others rely on different techniques like optical pumping /15/.

Another fascinating system is spin polarized hydrogen (H+), which is discussed in many of the articles in this journal issue. Compared to ³Het, spin polarized hydrogen offers several more exciting characteristics : Bose condensation in a dilute gas, no liquefaction at P = T = 0, etc.. Generally speaking, H atoms are lighter than ³He atoms so that quantum effects are more spectacular. Also, several isotopes can be studied : spin polarized Deuterium is also predicted to be a fascinating system, obeying Fermi statistics. Nevertheless, with H+ and D⁺, it seems impossible to vary continuously the polarization of the spins, which has to be practically complete for the system to be stable. Consequently, in this case X can not play the role of an additional thermodynamical variable. Another difference is that, in ³He, all macroscopic effects of the nuclear polarization are purely statistical effects, without any change of the interatomic potential ; the stability of the system is not critical and ones does not have to worry about atomic recombination or possible chain reactions. The surface (and bulk) relaxation problems are much less severe with ³He⁺ than H⁺, since nuclear magnetic moments are roughly 10³ times weaker than electronic spin moments. On the other hand, this is clearly a disadvantage if one intends to produce a spin polarized system by the so called "brute force" method.

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