

# Dynamic polarization of liquid $^3\text{He}$ at high temperatures

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The possibility of achieving a dynamic polarization of  $^3\text{He}$  nuclei by a method involving a spin resonance of electrons or  $\text{He}^+$  ions in the interior of the liquid or compressed by an electric field below its surface is analyzed. The electrons and  $\text{He}^+$  ions act as paramagnetic impurities, through which angular momentum of the electromagnetic field is pumped into the spin subsystem of  $^3\text{He}$  nuclei. The properties of polarized  $^3\text{He}$  are predicted at high temperatures, at which particle-statistics effects are small, but the liquid is a quantum liquid.

1. The static method<sup>1,2</sup> for polarizing  $^3\text{He}$  involves a rapid melting of polarized solid  $^3\text{He}$  by a strong magnetic field  $H$ . This method is used at low temperatures,  $T < 2\mu_n H$ , i.e., where the magnetic field  $H$  is capable of aligning the nuclear spins in the solid phase of  $^3\text{He}$ . Since the nuclear magnetic moment  $\mu_n$  is small, this can be done at  $T < 10^{-2}$  K at the magnetic fields  $H$  available today.

There is also a dynamic method for transferring polarization from electrons of a metal to liquid  $^3\text{He}$  at saturation of electron spin resonance<sup>3,4</sup> (ESR). The skin effect restricts the use of the ESR method to the case of small metal particles immersed in liquid helium. Even if a significant polarization of a liquid is achieved, the properties of the liquid remain difficult to study, since it is necessary to deal with a complex entity: a metal, which is coated with a layer of solid  $^3\text{He}$ , plus polarized liquid  $^3\text{He}$  ( $^3\text{He}_l$ ). The lifetime of  $^3\text{He}_l$  is very short in this case, since it is determined by a relaxation of nuclear spins at the solid surface of the vessel, i.e., ultimately by those metal particles which imparted the polarization to the liquid.

2. In this letter we wish to propose a dynamic method for polarizing  $^3\text{He}$  based on an ESR technique. The polarization would be caused in this method by electrons in direct contact with the liquid. There are two types of such electrons: "free" electrons and electrons of partially filled shells of positive ions, e.g.,  $\text{He}^+$ . Even in a weak magnetic field  $H \approx 1$  T, it is possible to cause a significant shift of the spin levels of an electron,  $2\mu_e H \approx 1$  K, where  $\mu_e$  is the magnetic moment of an electron ( $\mu_e \approx 10^3 \mu_n$ ). It is thus a simple matter to saturate the ESR at comparatively high temperatures,  $T < 2\mu_e H$ .

According to reliable arguments,<sup>5</sup> the electrons and the  $\text{He}^+$  ions in liquid helium are quite different entities. The electron creates a spherical cavity of radius  $r_- \approx 20$  Å in the liquid. This cavity has an effective mass  $\approx 300$  times the mass of the He atom.

The positive  $\text{He}^+$  ion, in contrast, is surrounded by a sphere of solid He, with a

size  $r_+ \approx 10 \text{ \AA}$  and an effective mass  $\approx 100$  times the mass of the He atom. Such large effective masses of the charges in liquid He mean that the electrons and  $\text{He}^+$  ions can be treated as paramagnetic impurities. The interaction of the spin of such an impurity,  $\sigma_1$ , with the nuclear spin  $\sigma_2$  is determined by the sum of the dipole-dipole component ( $V_1$ ) and the contact component ( $V_2$ ):

$$V = V_1 + V_2, \quad V_2 = -\frac{8\pi}{3} \mu_d \mu_n (\sigma_1 \cdot \sigma_2) \delta(\mathbf{r}),$$

$$V_1 = \mu_d \mu_n \left\{ \frac{(\sigma_1 \cdot \alpha_2)}{r^3} - 3 \frac{(\sigma_1 \cdot \mathbf{r})(\sigma_2 \cdot \mathbf{r})}{r^5} \right\}, \quad (1)$$

where  $r$  is the distance between the electron and the  $^3\text{He}$  nucleus. In an ESR case, the electrons are characterized by two spin levels,  $\epsilon_1$  and  $\epsilon_2$ :  $\epsilon_1 - \epsilon_2 = 2\mu_e H$ . The liquid  $^3\text{He}$  is a nonequilibrium mixture of two spin components with chemical potentials  $\mu_1$  and  $\mu_2$ . When the spins  $\sigma_1$  and  $\sigma_2$  flip simultaneously (flip-flops), the liquid acquires an energy  $\epsilon_1 - \epsilon_2$  from the electron. The "extra" energy  $\epsilon_1 - \epsilon_2 - \mu_1 + \mu_2$  goes into exciting the liquid in this case (the excitations are collective and one-particle excitations). The inverse process also occurs: The nucleus flips the electron spin, which acquires the deficient energy  $\epsilon_1 - \epsilon_2 - \mu_1 + \mu_2$  from the liquid. Such a process is less probable than the forward process at a low temperature  $T < 2\mu_e H$ , since it involves an absorption of real thermal excitations, whose density is negligibly low. In a strong magnetic field ( $T \ll \epsilon_1 - \epsilon_2 = 2\mu_e H$ ) the electrons polarize the liquid until the difference  $\mu_1 - \mu_2$  becomes comparable to  $\epsilon_1 - \epsilon_2 = 2\mu_e H$ . We will show that this limitation is not important and that a complete polarization of  $^3\text{He}$  can be achieved even in comparatively weak magnetic fields.

3. A fundamental distinction between liquid  $^3\text{He}$  and an ideal gas is that the degeneracy temperature  $T_F$  of the former is anomalously low. This temperature is a characteristic exchange energy; it determines the temperature dependence of the magnetic susceptibility,  $\chi(T)$ :

$$\chi(T) \approx \frac{C}{T} \text{ at } T \gg T_F, \quad \chi(T) \approx \frac{C}{T_F} \text{ at } T \ll T_F. \quad (2)$$

At the equilibrium density of  $^3\text{He}$ , the parameter  $T_F$  is an order of magnitude smaller than the gas value  $T_F^0 \approx 3 \text{ K}$ . As the density of the liquid increases, this parameter decreases rapidly. Another energy scale of the liquid, in this case a very large one (13–20 K), and one which is insensitive to the type of statistics of the particles, is the energy of the zero-point vibrations of the liquid,  $\bar{K}_0$ . This energy is a strong increasing function of the density of the  $^3\text{He}$ . This parameter is determined by the average kinetic energy of a  $^3\text{He}$  atom at  $T=0$ :

$$\bar{K}(T) \approx \frac{3}{2} T \text{ at } T \gg \bar{K}_0, \quad \bar{K}(T) \approx \bar{K}_0 \text{ at } T \ll \bar{K}_0. \quad (3)$$

The existence of the very broad temperature interval  $T_F \ll T \ll \bar{K}_0$  provides a unique opportunity to polarize the  $^3\text{He}$  completely at high values of  $T$ , at which the  $^3\text{He}$  is a quantum but nondegenerate liquid.<sup>3</sup> The difference between chemical poten-

tials  $\mu_{\uparrow} - \mu_{\downarrow}$  is at a maximum when the liquid is completely polarized, and it has a scale value of  $T_F$  (Ref. 1):  $\mu_{\uparrow} - \mu_{\downarrow} \approx 2T_F$ . Accordingly, even in a field  $H > 2 T$  this temperature interval can easily be reached:

$$\mu_{\uparrow} - \mu_{\downarrow} \approx 2T_F < T < 2\mu_e H \ll \bar{K}_0.$$

In a first approximation, we can set  $T_F = 0$  at such values of  $T$ , and the basic thermodynamic and kinetic characteristics of  ${}^3\text{He}_{\uparrow}$  and  ${}^3\text{He}$  will differ only slightly. In addition, they are related by a scaling of the temperature by a factor of 3/4 to the corresponding characteristics of liquid nonsuperfluid  ${}^4\text{He}$  (Ref. 6). An exceptional case is the entropy  $S$  of  ${}^3\text{He}$ , which at  $T \gg T_F$  has a spin component of  $\ln 2$  per atom. This component is not present in the entropy ( $S_{\uparrow}$ ) of completely polarized  ${}^3\text{He}$ :

$$S(T) - S_{\uparrow}(T) = \ln 2 \text{ at } T \gg T_F.$$

4. Let us analyze the contributions of the dipole-dipole ( $V_1$ ) and contact ( $V_2$ ) forces in (1) to the polarization of  ${}^3\text{He}$  for the case of an electron bubble. The energy at which the electron enters liquid helium is very large ( $\approx 1$  eV), so in a first approximation there is no region in which the electron wave function  $\psi(r)$  in the bubble and the density of the liquid  $n(r)$  are both nonzero:  $\psi(r) = 0$  at  $r > r_-$ , and  $n(r) = 0$  at  $r < r_-$ . In this approximation, the contact forces in (1) thus do not contribute to the polarization of the liquid. Their primary contribution is associated with the polarization of the  ${}^3\text{He}$  vapor which fills the electron bubble. Since the density of the vapor depends very strongly (exponentially) on the temperature at  $T < 2$  K, dipole-dipole forces makes the predominant contribution at such low values of  $T$ . For a dilute solution of  ${}^3\text{He}$  in  ${}^4\text{He}$ , however, the situation is different, because of the pronounced enrichment of the surface of the electron bubble with  ${}^3\text{He}$  atoms.<sup>7</sup> If we take into account the blurring of the boundary separating the electron and the liquid, we find that contact forces can dominate the polarization of a dilute solution of  ${}^3\text{He}$  in  ${}^4\text{He}$ .

The contributions  $V_1$  and  $V_2$  in (1) to the polarization of  ${}^3\text{He}$  can be separated experimentally, and accurately, in the case of the  $\text{He}^+$  ion. In pure  ${}^3\text{He}$ , the contact forces  $V_2$  can take part in the polarization of the liquid indirectly: First, an electron polarizes the solid crust surrounding the ion. Then, through a direct exchange of atoms (solid  $\rightarrow$  liquid  ${}^3\text{He}$ ), the polarization is transferred to the liquid. In a  ${}^3\text{He}$ - ${}^4\text{He}$  solution, however, a positive  $\text{He}^+$  ion is surrounded by a crust of pure  ${}^4\text{He}$  (Ref. 5), so the contact forces are "turned off," and only the dipole-dipole forces operate. Since a  ${}^3\text{He}$ - ${}^4\text{He}$  solution does not stratify at  $T \sim 1$  K, even at a low average concentration of  ${}^4\text{He}$  in liquid  ${}^3\text{He}$ , all the  ${}^4\text{He}$  collects near  $\text{He}^+$  ions.

5. The time scale ( $T_0$ ) for electron spin flip in liquid  ${}^3\text{He}$  is<sup>8</sup> on the order of  $10^{-3}$  s. This time has not been measured for the  $\text{He}^+$  ion, but it is shorter at low temperatures  $T < 2$  K, at which contact forces need not be considered. According to a dimensionality estimate, the probability for dipole-dipole flip of the spin of an electron at ESR is proportional to  $r_0^{-6} f(pr_0)$ , where  $p$  is a characteristic momentum of the liquid, and  $r_0$  is the radius of the bubble (for the electron) or the radius of the solid sphere (for the  $\text{He}^+$  ion). The exact dependence of  $f$  on the parameter  $pr_0$  is not important for our purposes. There is, however, an important point: For helium the condition  $pr_0 \gg 1$  holds, and a semiclassical estimate of the function  $f$  indicates that

it falls off rapidly with increasing  $pr_0$ . At the equilibrium  $^3\text{He}$  density, the parameter  $r_+$  (for an ion) is smaller by a factor of 2 or 3 than  $r_-$  (for an electron). We thus have a basis for an optimistic estimate of the time scale for electron spin flip of  $\text{He}^+$ :  $\sim 10^{-5}$  s. Since the size of the electron bubble shrinks rapidly with increasing density of the liquid helium,<sup>5</sup> this is also a reasonable estimate of the time scale for electron spin flip at high pressures ( $\sim 30$  atm).

Over a time of 1 s at ESR, one electron can thus flip the spins of  $10^5$   $^3\text{He}$  nuclei. However, there is a "leakage" of the polarization of the liquid because of weak dipole-dipole nuclear forces, which differ from  $V_1$  in (1) by the small factor  $\mu_n/\mu_e \approx 10^{-3}$ . The time scale ( $T_1$ ) for the relaxation of liquid  $^3\text{He}$  due to these forces has been measured<sup>9</sup> for a liquid polarized by a weak magnetic field at  $T \sim 1$  K; it is on the order of  $10^3$  s. It is thus possible to achieve a significant polarization of  $^3\text{He}$  if the charge density  $\rho$  is higher than the density of the liquid,  $n$ , reduced by the time ratio  $T_0/T_1$ :  $\rho > 10^{14}$  cm<sup>3</sup>. Such a high number density of charges in helium could not be arranged. However, an experiment could be carried out with a  $^3\text{He}$  film of thickness  $d < 10^{-5}$  cm on which there is a surface charge density  $\approx 10^9$  cm<sup>2</sup>. This is a completely feasible density of charges compressed by an electric field below the surface of liquid helium.

The method which we have proposed for polarizing  $^3\text{He}$  allows an "instantaneous removal" of charges from the liquid. Here it is sufficient to replace the compressing electric field by a field which squeezes charges away from the surface of the helium film. In this case one can study the natural relaxation of a highly polarized film of liquid  $^3\text{He}$ .

6. It would also be interesting to measure the lifetime of  $^3\text{He}$ , in the presence of electrons or  $\text{He}^+$  ions. If such measurements were carried out without a magnetic field  $H$ , we would have the standard situation of a relaxation of nuclear spins due to paramagnetic impurities, with the polarization of the nuclei being transferred back to the electrons. In a strong magnetic field,  $H$ , however, the spins of the electrons are frozen, and inverse flip-flops are forbidden. The relaxation of nuclear spins by electrons is of purely dipole-dipole origin in this case, since the  $V_1$  interaction in (1) does not conserve the total spin of the system consisting of the electron plus the  $^3\text{He}$  nucleus. There is the possibility of a process in which the spin of the nucleus is flipped, while that of the electron is not. Since the  $V_2$  contact forces in (1) do conserve the total spin, a measurement of nuclear relaxation with and without a field  $H$  would make it possible to distinguish the contributions of  $V_1$  and  $V_2$  not only to the polarization of liquid  $^3\text{He}$  but also to its relaxation.

We note in conclusion that the polarization method proposed here also applies to solid  $^3\text{He}$ . In the limit of a strong magnetic field,  $H \gg \bar{K}_0$  ( $H > 20$  T), the difference between liquid and solid helium disappears in an experiment involving the ESR of charges in  $^3\text{He}$ , since the electron transfers such a large amount of energy to the condensed medium in the course of flip-flops that an atom whose nuclear spin has flipped moves with an energy  $\approx 2\mu_e H \gg \bar{K}_0$ ; i.e., it is free. The electrons and  $^3\text{He}$  ions "shoot out" fast, polarized  $^3\text{He}$  atoms.

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