Paramagnetic relaxation of negative ions in a degenerate ^3He–^4He solution

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The spin relaxation of negative ions in a degenerate ^3He–^4He solution and also in pure ^3He is analyzed. Electron spin flips result primarily from the dipole-dipole interaction with the magnetic moments of the ^3He atoms. The system is modeled as a large bubble filled with an ideal Fermi gas to find the longitudinal relaxation time as a function of the dimensions of the bubble, the temperature, and the external magnetic field. The results derived give a quantitative description of relaxation in degenerate ^3He–^4He solutions under the inequalities $a > k_F^{-1} > f/\pi$, where a is the bubble radius, k_F is the momentum of the quasiparticles, and f is the amplitude for scattering of quasiparticles by each other. The use of the results for a qualitative description of relaxation in pure ^3He is discussed.

I. INTRODUCTION

An electron which enters liquid helium from outside is known to be repelled by the atoms of the liquid. It forms a spherical cavity around itself with a radius of 10–20 Å, depending on the external pressure.[1,2] In its ground state in the bubble, the electron is described by an s-state wave function. An external magnetic field lifts the Kramers degeneracy and causes spin splitting of the ground state into two levels. Transitions between these levels can be studied by electron spin resonance.

The active experimental and theoretical research on negative ions has been, and still remains, aimed primarily at determining the transport and optical properties of these ions. There are two basic factors which obstruct paramagnetic-resonance experiments. In the first place, the spin relaxation time would have to be rather long because of the weak magnetic interaction between an electron in the bubble and the atoms of the liquid. Second, space-charge limitations would keep the attainable ion densities at a very low level. Recently, these difficulties were overcome, and observations of ESR in liquid ^4He and ^3He at temperatures $T > 1.4$ K and ion densities $n \sim 10^{10}$ cm^{-3} have been reported.[3,4]

At these temperatures, the degeneracy has only a minor influence on the ^3He, and the results of Ref. 4 can be interpreted at a qualitative level in classical theory.[5] In the present paper we are concerned with low temperatures, $T < 0.1$ K, at which liquid ^3He is a Fermi liquid, the mean free path of a quasiparticle is far greater than the dimensions of a bubble, and a quantum-mechanical approach must be taken in calculations on paramagnetic relaxation. The relaxation of an electron spin results from a scattering of a quasiparticle by the electron, by analogy with the relaxation of a paramagnetic impurity in a metal. Since distances on the order of the dimensions of the bubble are important, as we will see below, it is necessary to allow for the distortions of the wave function of a quasiparticle by the potential field of the bubble despite the small wavelength of the quasiparticle, $\lambda \sim p_F^{-1}$.

2. HAMILTONIAN OF THE MAGNETIC INTERACTION

The energy of the magnetic moment ($\boldsymbol{\mu}_e = -2\mu_B \mathbf{s}$) of an electron in the field set up by the magnetic moment ($\boldsymbol{\mu} = 2\mu_n \mathbf{I}$) of the nucleus of a ^3He atom is

$$U(\mathbf{r}) = 2\mu_B s^i \mu^j (3\hat{r}_i\hat{r}_j - \delta_{ij}) r^{-3} + (16\pi/3)\mu_B \boldsymbol{\mu}\mathbf{s}\delta(\mathbf{r}), \qquad (1)$$

where the operator $\mathbf{I}$ represents the nuclear spin, μ_n is the nuclear magneton, the operator $\mathbf{s}$ represents the electron, and $\mu_B = 0.9\cdot 10^{-20}$ erg/G is the Bohr magneton. The first term in (1) gives us the dipole-dipole interaction energy, while the second gives the energy of the contact interaction. We will show that this contact interaction can be ignored.

The depth to which an electron with a kinetic energy $E_0 \sim \hbar^2/ma^2$ (a is the bubble radius) penetrates under a barrier of height $U_0 \approx 1$ eV is given by $l_e = \hbar/[2m(U_0 - E_0)]^{1/2}$. This depth is small, 2–3 Å. The region over which the bubble wall is spread is the same size.[6] For the dipole-dipole interaction, the important regions are within a distance are on the order of the bubble radius, $a > l_e$, from the surface of the bubble so that the wall thickness may be ignored. We average the first term in (1) over the wave function of the ground state of an electron in a spherical well with an inpenetrable wall[7]:

$$\varphi_0^2(r) = a^{-3} f\left(\frac{r}{a}\right), \quad f(x) = \frac{1}{2\pi}\left(\frac{\sin \pi x}{x}\right)^2, \quad E_0 = \frac{\hbar^2\pi^2}{2ma^2}. \qquad (2)$$

At $R > a$ we find

$$U_d(\mathbf{R}) = 6\mu_B s^i \mu^j (\hat{R}_i\hat{R}_j - \delta_{ij}/3) R^{-3}. \qquad (3)$$

This expression should be compared with the expectation value of the second term in (1):

$$U_c(\mathbf{R}) = 16\pi\mu_B \mathbf{s}\boldsymbol{\mu}\varphi^2(\mathbf{R})/3. \qquad (4)$$

In the model of a square well of depth U_0, the wave function of an electron at the bubble boundary would be[7]

$$\varphi^2(a) \approx E_0/2\pi a^3 U_0, \qquad (5)$$

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from which we conclude that the ratio of the energies is small even at the boundary:

$$U_c(a)/U_d(a)\approx 4E_0/9U_0 \ll 1. \qquad (6)$$

Furthermore, the contact interaction is concentrated in the region in which the wave function of the electron penetrates into the liquid, $\sim l_e$, while the dipole-dipole interaction is concentrated in a region with a size of order $a > l_e$. The net result of these two circumstances is that the U_c contribution during the relaxation is more than two orders of magnitude smaller than the U_d contribution.

The Hamiltonian of the interaction of the electron and nuclear moments can thus be written

$$V_{\alpha\sigma|\alpha'\sigma'}(\mathbf{r}) = 4\mu_B\mu_n s_{\alpha\alpha'}{}^i I_{\sigma\sigma'}{}^j (3\hat{r}^i\hat{r}^j - \delta^{ij}) r^{-3}. \qquad (7)$$

We will use the same Hamiltonian to describe the interaction of the quasiparticles with an electron. Since the paramagnetic resonance frequency is far lower than the frequencies of optical transitions of an electron in a bubble, we can ignore the effect of the absorbed radiation on the orbital motion of the electron. Going over to the second-quantized representation, we then find

$$\mathcal{H}_{int} = \sum_{\alpha\sigma\mathbf{k}\alpha'\sigma'\mathbf{k}'} c_\sigma^+ a_{\mathbf{k},\alpha}^+ \langle \mathbf{k};\sigma,\alpha|V|\mathbf{k}';\sigma',\alpha'\rangle a_{\mathbf{k}'\alpha'} c_{\sigma'},$$
$$\langle \mathbf{k};\sigma,\alpha|V|\mathbf{k}';\sigma',\alpha'\rangle = \int d^3r\, \psi_{\mathbf{k}}^*(\mathbf{r}) V_{\alpha\sigma|\alpha'\sigma'}(\mathbf{r}) \psi_{\mathbf{k}'}(\mathbf{r}), \qquad (8)$$

where $\psi_{\mathbf{k}}(\mathbf{r})$ is the complete set of "in" states (or "out" states) for the scattering of the quasiparticles in the potential field of the bubble,[8,9] the operator $a_{\mathbf{k}\alpha}^+$ creates a quasiparticle in the corresponding state, and the operator c_σ^+ creates an electron in the ground state $\varphi_0(\mathbf{r})$ with a spin projection σ.

3. CALCULATION OF THE RELAXATION TIME

If we ignore the recoil of the ion in a collision with a quasiparticle, we can treat the ions as a set of randomly located paramagnetic centers. The gyromagnetic ratio for an electron is far larger than that for the ^{3}He nucleus, so it can be assumed that a static magnetic field $\mathbf{H}$ acts only on the electron spin. Since the expected relaxation time is rather long, it is clear that under the experimental conditions the width of the paramagnetic resonance will be determined not by the transverse relaxation time T_2 but by the variations in the magnetic field, so that we need calculate only the longitudinal relaxation time T_1. The time T_1 is expressed in the usual way, with the help of the population balance equations, in terms of the probability $W_{1\to 2}$, for a transition of an electron from state 1 (with a spin directed along the field) to state 2 (spin directed opposite the field) as a result of a collision with a quasiparticle, and in terms of $W_{2\to 1}$, the probability for the inverse transition:

$$T_1^{-1} = W_{1\to2} + W_{2\to1}, \qquad (9)$$

where

$$W_{\substack{1\to2\\2\to1}} = \frac{2\pi}{\hbar}\sum_{\alpha\beta}\int d^3k_1\, d^3k_2\, n_F(\xi_{k_1})$$
$$\times[1-n_F(\xi_{k_2})]\delta(\pm\hbar\omega_0+\xi_{k_1}-\xi_{k_2})\,|\langle \mathbf{k}_1;1,\alpha|V|\mathbf{k}_2;2,\beta\rangle|^2, \qquad (10)$$

and $\omega_0 = 2\mu_B H$ is the paramagnetic resonance frequency. It has been assumed here that the liquid is in thermodynamic equilibrium and serves as a heat reservoir for the ions.

An important point is that in calculating T_1 we cannot use the method[10] of approximating the two-point correlation function which figures in (10) by the contributions from two classical paths of the quasiparticle: a straight path and one with a specular reflection from the bubble. The diffraction region makes a contribution comparable to that of the region in which the geometric-optics approximation is valid. We should therefore calculate the transition matrix elements directly, making use of the exact states for the scattering of a quasiparticle by an inpenetrable sphere of large radius.

We describe the incident particles by the functions $\psi_{\mathbf{k}}^{(+)}$ and the scattered particles by the functions $\psi_{\mathbf{k}}^{(-)}$; the notation is that of Refs. 7 and 9. We assume

$$D^{pq}(\mathbf{k}_2,\mathbf{k}_1) = \int d^3r\, \psi_{\mathbf{k}_2}^{(-)*}(\mathbf{r})\,(\hat{r}^p\hat{r}^q - \delta^{pq}/3) r^{-3} \psi_{\mathbf{k}_1}(\mathbf{r}). \qquad (11)$$

To calculate the probability for the scattering of a quasiparticle by a bubble accompanied by a flip of the electron spin from state σ_1 to state σ_2, we will need the quantity

$$M_{\sigma_1\sigma_2} = \int d\hat{k}_1\, d\hat{k}_2 D^{pq}(\mathbf{k}_2,\mathbf{k}_1) D^{p'q'*}(\mathbf{k}_2,\mathbf{k}_1)\,\mathrm{Tr}(s^q s^{q'})$$
$$\times \mathrm{Tr}(I^p\hat{P}_1 I^{p'}\hat{P}_2), \qquad (12)$$

where the operators $\hat{P}_{1,2}$ project onto states σ_1 and σ_2. In the case $\sigma_1 = 1$, $\sigma_2 = 2$, the product of the traces in (12) is

$$\delta^{qq'}(\delta^{pp'} - \delta^{p3}\delta^{p'3} - ie^{pp'3})/8. \qquad (13)$$

Now substituting the expansion[8,9]

$$\psi_{\mathbf{k}}^{(\pm)}(\mathbf{r}) = (2k)^{-1}\sum_l i^l(2l+1)e^{\pm i\delta_l(k)} R_{kl}(r) P_l(\hat{\mathbf{k}}\hat{\mathbf{r}}) \qquad (14)$$

into (11), introducing the radial matrix element

$$(k_2, m|r^{-3}|k_1, l) = \int_0^\infty R_{k_2m}(r) r^{-1} R_{k_1l}(r)\, dr \qquad (15)$$

and assuming $l \sim k_F a \gg 1$, we find

$$M_{21} = (2\pi^2/3k_1k_2)^2 \sum_l l\{|(k_2,l|r^{-3}|k_1,l)|^2$$
$$+ {}^3/_2|(k_2,l+2|r^{-3}|k_1,l)|^2 + {}^3/_2|(k_2,l-2|r^{-3}|k_1,l)|^2\}. \qquad (16)$$

The radial matrix elements can be calculated in the semiclassical approximation, in view of the large radius of the ion $k_F a \gg 1$. The radial motion of quasiparticles with an energy near ε_F occurs in the potential

$$U(r) = U_0\theta(a-r) + \frac{\hbar^2}{2m_s}\frac{(l+{}^1/_2)^2}{r^2}, \qquad (17)$$

which has a stopping point $r_0 = (l+1/2)/k > a$ in the case $l > ka$. At $r < r_0$, the function $R_{kl}(r)$ decays rapidly and does not contribute to integral (15) in the leading order in $k_F a$. In the classically allowed region, $r > r_0$, the function $R_{kl}(r)$ oscillates with a period k_F^{-1}: For[9] $l < ka$ we have

$$R_{kl} \approx \frac{(4k/r)^{1/2}}{[(kr)^2 - l^2]^{1/4}}\sin[\alpha_l(r) + \delta_l], \quad \delta_l = -\alpha_l(a),$$
$$\alpha_l = kr[(1-t^2)^{1/2} + t\arcsin t - \pi t/2], \quad t = l/kr, \qquad (18)$$

while for $l > ka$ we have the semiclassical phase[1)] $\delta_l = 0$. Using (18) for the radial integrals, we can derive the expressions

$$I_l = \int dr\, r^{-1} R_{kl}^2(r)$$
$$= \theta(l-ka)\, 2k^2/l^2 + \theta(ka-l)\, \{1-[1-(l/ka)^2]^{1/2}\}\, 2k^2/l^2, \tag{19}$$
$$J_l = \int dr R_{k,l+1}(r)\, r^{-1} R_{k,l-1}(r)$$
$$= \theta(l-ka)\, 2k^2/3l^2 + \theta(ka-l)\, \frac{1+2[1-(l/ka)^2]^{1/2}}{1+[1-(l/ka)^2]^{1/2}} \frac{2}{3a^2}.$$

Since partial waves with large angular momenta $l \sim k_F a$ are important, we can replace the sums by integrals in (16):

$$\int_0^\infty l I_l^2\, dl = \frac{4k^2 \ln 2}{a^2}, \quad \int_0^\infty l J_l^2\, dl = \frac{2k^2(5\ln 4 - 4)}{9a^2}. \tag{20}$$

Also using the expression

$$\Gamma(\omega, T) = \int dE n_F(E)\, [1 - n_F(E+\hbar\omega)]$$
$$= \hbar\omega [1 - \exp(-\hbar\omega/T)]^{-1}, \tag{21}$$

we find the following result for the longitudinal relaxation time:

$$\hbar T_1^{-1} = [\Gamma(\omega_0, T) + \Gamma(-\omega_0, T)]\, (\mu_B \mu_n m_3/\hbar^2 a)^2 \cdot \times 2^5 (\ln 4 - 0.5)/3\pi. \tag{22}$$

An important point is that the expression which we have derived for the relaxation time does not depend on the Fermi momentum of the quasiparticles or thus on the density of the Fermi gas. The momentum k_F enters the problem twice: First, it enters through the state density of quasiparticles, $\rho = m_3 k_F / 2\pi^2$. Second, the relation between the quasiparticle wavelength k_F^{-1} and the bubble radius determines the nature of the scattering states $\psi_{\mathbf{k}}^{(\pm)}$ and thus the size of the matrix element from the magnetic interaction V in (10). In the limit of a large bubble radius $k_F a > 1$, the dependence on k_F cancels out. This distinguishes our problem from a calculation of the relaxation time of a short-range paramagnetic impurity in a metal. It also demonstrates a need for a careful account of the distortion of the quasiparticle plane waves by the potential field of the bubble.

Under the same conditions, the ion mobility μ depends strongly on the density of the Fermi gas[8]: $\mu \sim k_F^{-4}$. The difference between T_1 and μ arises from the different structures of the matrix elements. The mobility contains, instead of the matrix elements of the long-range operator respresenting the dipole-dipole interaction, given by (7), matrix elements of the short-range operator representing the force $\nabla U_0(r)$, which is exerted on a quasiparticle by an ion.[8]

The very same factor which leads to the cancellation of k_F in (22) dictates the dependence of the relaxation time on the radius a. In a classical description of the relaxation, valid at high temperatures, the results $T_1 \sim a^3$ is found.[4]

4. CONCLUSION

These calculations have dealt with the case of an ideal Fermi gas. We will thus first consider the spin relaxation of negative ions in a degenerate ^3He–^4He solution with a ^3He concentration c. We assume that the conditions are such that the following relations hold:

$$a \gg k_F^{-1} \gg f/\pi, \tag{23}$$

where f is the amplitude for the scattering of quasiparticles by each other. The left side of this inequality means that the bubble size is far greater than the average distance between ^3He atoms. At a concentration $c = 6\%$ we have $k_F = k_F^0 c^{1/3}$, where $k_F^0 = 0.8\ \text{Å}^{-1}$ is the Fermi momentum of pure ^3He. When the external pressure vanishes, with $a = 19$ Å, the basic inequality $k_F a = 5.6 \gg 1$ holds with an ample margin. The right side of inequality (23) means that the gas of quasiparticles can be treated as ideal. Under these conditions, the criterion for an ideal gas,[11–13]

$$k_F f/\pi = 0.15 \ll 1, \tag{24}$$

where we have set[14] $f = -1.5$ Å, is satisfied to roughly the same extent.

In addition to the potential of the inpenetrable sphere, a polarization potential acts on a quasiparticle[15]:

$$U_{pol}(r) = -(\alpha e^2/2r^4)\,(1 - v_3/v_4), \tag{25}$$

where α is the polarizability of the ^3He atom, and $v_{3,4}$ are the volumes per ^3He atom and ^4He atom in solution. Since we have $v_3/v_4 = 1.27$ the potential U_{pol} creates a weak repulsion; the small value of the ratio $U_{pol}(a)/\varepsilon_F = 0.1$ means that we can ignore this repulsion.

On occasion, by analogy with a free ^4He surface,[16] there has been a discussion of the possible formation of a layer of ^3He atoms on the surface of a bubble, due to the presence of a bound state. Since a quasiparticle is repelled from an ion at distances from the boundary greater than the atomic size, according to (25), such a layer—if it exists—could have a thickness only on the atomic scale. In nonmagnetic potential scattering of free quasiparticles, the presence of a layer would cause an unimportant renormalization of the radius a. Furthermore, there would be the possiblity of exchange scattering, so that the mobility might grow logarithmically with decreasing temperature.[9] Spin relaxation of an electron in the bubble is caused by the oscillating magnetic field of the quasiparticles which are being scattered, so that the presence of a layer would have an effect on the relaxation time only if the exchange scattering were fairly strong. Incidently, the hypothesized existence of a bound state of a quasiparticle at the surface of a bubble has yet to receive adequate experimental verification.

In pure ^3He, the interparticle intractions cause not only renormalization of the mass of the quasiparticles but also (first) means that the field exerted on the quasiparticles by the bubble no longer reduces just to the potential of an inpenetrable sphere and (second) gives rise to a final-state interaction, i.e., an interaction between the incident and scattered waves. Consequently, the calculation which has been carried out is only qualitatively applicable to ^3He, except for the dependence on the temperature and the magnetic field, which is purely a consequence of the Fermi statistics of the quasiparticles. Since the relaxation time is independent of k_F in the ideal-gas model, at $k_F a > 1$, the ratio of the relaxation times in a 6% solution of ^{3}He and also in pure ^{3}He will be determined exclusively by the ratio of the effective masses of the quasiparticles, to the extent that the ideal-gas model is valid for liquid ^{3}He. Here the product $m_3^2 T_1$ must be approximately the same. The pronounced increase in the relaxation time in a solution should occur at that dilution at which the wavelength of the quasiparticle becomes comparable to the radius of an ion, $k_F \sim a$. The general approach of the calculations remains valid in this case, but the sums over partial waves in (16) can no longer be replaced by integrals, and in evaluating radial matrix elements (15) we can no longer use semiclassical expressions for the radial wave functions.

An extrapolation of (22) to the experimental conditions of Ref. 4, with $T = 1.4$ K, yields $T_1 < 0.1$ s and agrees qualitatively with the experimental data. In the case of degeneracy of pure ^{3}He we would expect $T_1 \gtrsim 1$ s. As the magnetic field increases, T_1 decreases. The reason is that as an electron undergoes a transition from the upper spin state to the lower one with increasing field, the phase volume of the quasiparticle excited in the process increases.

With a further decrease in the temperature, the relaxation time increases, but it may turn out that in the super fluid phase this time will again be rather short, if the magnetic field is chosen to satisfy the condition $\hbar\omega_0 = 2\Delta$. This circumstance should result from a resonance of the ESR frequency with the creating energy of a pair of quasiparticles undr conditions such that the state density has a singularity at the Fermi surface. Interestingly, such a situation might in principle be achieved in helium, by virtue of the strong inequality $\mu_B > \mu_n$, while in ordinary superconductors magnetic fields at this level would destroy the superconductivity.

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[1)]There is a misprint in Eq. (A5) in Ref. 9: The function arcsin should appear instead of arctg (arctan) in α_l.

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