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Richard A. Webb University of South Carolina - Columbia, webbra@mailbox.sc.edu

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### Ferromagneticlike Resonance Behavior in Superfluid <sup>3</sup>He-B

#### R. A. Webb

Argonne National Laboratory, Argonne, Illinois 60439 (Received 29 June 1977)

Nonlinear cw NMR behavior, similar to that observed on ferromagnets at high rf powers, has been observed in superfluid  ${}^{3}He-B$  using a superconducting quantum interference device (SQUID) magnetometer in fields of 31, 102, 180, and 308 Oe and at a pressure of 21 bar. It is observed that beyond some threshold value of the transverse  $H_1$  field the z component of magnetization anomalously decreases and is followed by a rapid increase back to the normal resonance curve as the frequency of the  $H_1$  field is swept through resonance.

Nonlinear cw NMR behavior has been observed in superfluid  ${}^{3}$ He-B at a pressure of 21 bar using a superconducting quantum interference device (SQUID) in measuring fields of 31, 102, 180, and 308 Oe. SQUID cw NMR directly measures the decrease in the z component of magnetization,  $M_{\bullet}$ , as the frequency of the rf field  $\vec{H}_{1}$ , perpendicular to the static field  $\vec{H}_0$ , is slowly swept through resonance. The change in  $M<sub>s</sub>$  is found to obey the usual Bloch equation for  $H_1$  fields below some threshold value. Above this threshold, the nonlinear behavior manifests itself in an abnormal decrease of  $M<sub>e</sub>$  followed by a sudden increase of  $M<sub>s</sub>$  back to the normal resonance curve. Both the magnitude of the jump and the frequency at which it occurs depend upon  $H_1$ ,  $T/T_c$ , and  $H_0$ . The value of the  $H_1$  field at which these nonlinear effects first occur depends both upon  $T/T_c$  and  $H_0$ . A similar nonlinear effect has been observed by Osheroff' during measurements of the transverse absorption spectrum of  ${}^{3}$ He-B, although no detailed investigation of the phenomenon was attempted. This behavior is similar to the NMR results obtained on ferromagnets at high rf powers.' In the ferromagnetic resonance case it has been shown<sup>2, 3</sup> that these nonlinear effects arise from the coupling of spin-wave modes to the uniform precession of the magnetization vector.

The adiabatic demagnetization cell used for the experiments reported here has been described elsewhere.<sup>4</sup> The <sup>3</sup>He measured was contained in a B-mm-i. d. tower located above the main cerium magnesium nitrate cell. Absolute temperatures were determined with the aid of the La Jolla phase diagram. $5$  For the work reported here, the calibration of the SQUID magnetometer was determined using pulsed NMR and adiabatic fast passage at 15 mK. A complete description of the techniques of SQUID NMR has been given elsewhere.<sup>6</sup>

Some typical examples of the kinds of nonlinear events that were observed in a static field of 102 Oe are shown in Fig. 1 for a reduced temperature of  $T/T_c = 0.922$ . The sweep rate for all four traces was 0.42 kHz/sec. The first trace, obtained using  $H_1 = 2.49 \times 10^{-2}$  Oe, is an example of the normal superfluid cw NMR line. The twopeak resonance curve is a result of the inhomogeneity in  $\vec{H}_0$  over the sample volume and has the same shape in the normal phase. The measured separation between the two peaks was  $\Delta H = (0.016$  $\pm$  0.005) $H_0$  and is explained in detail elsewhere.<sup>6</sup> Increasing the  $H_1$  field a few percent to 2.54  $\times$  10<sup>-2</sup> Oe produces the first nonlinear event at this temperature and is shown in the second trace



FIG. 1. Typical cw NMR traces observed in  ${}^{3}$ He-B at a reduced temperature  $T/T_c = 0.922$  in 102 Oe at a pressure of 21 bar. The  $H_1$  field used to obtain the four traces was (1)  $2.49 \times 10^{-2}$  Oe; (2)  $2.54 \times 10^{-2}$  Oe; (3)  $3.5 \times 10^{-2}$  Oe; and (4)  $5.93 \times 10^{-2}$  Oe. The arrows indicate the direction of sweep.

in Fig. 1. The z component of magnetization initially decreases more than would be expected for this  $H_1$ , field. This is followed by a rapid and nearly discontinuous increase of the magnetization back to the normal resonance curve. This small nonlinear event appears suddenly and the magnitude of the jump slowly increases with increasing  $H_1$  field. Sweeping the frequency of the  $H<sub>1</sub>$  field in the reverse direction never produces a nonlinear event but yields the normal resonance curve. The third trace,  $H_1 = 3.5 \times 10^{-2}$  Oe, is an example of one large nonlinear event. The transition from a small jump to a large jump also occurs suddenly, although the value of the  $H_1$  field at which this transition first occurs is somewhat less reproducible than for the onset of the small jumps. The dashed line is the resonance curve followed when sweeping in the reverse direction. The nonlinear portion of the resonance curve is reversible so long as the jump in magnetization is not allowed to occur. Both the magnitude of the large jump and the frequency at which it occurs is nearly independent of the  $H_1$  field. However, as the  $H_1$  field is increased further, there is yet another sudden transition from one large jump to two large jumps. The fourth trace in Fig. 1, obtained for  $H_1 = 5.93 \times 10^{-2}$  Oe, is an example of the two large jumps and is the most frequently observed curve. The dotted curves again indicate the resonance curve followed when the direction of the frequency sweep is reversed. After going through the first jump, but before the second jump has occurred, one can reverse the direction of the sweep and follow a curve similar to that shown in the third trace. This second nonlinear curve is also quite stable. For example, one can sit at any point on the second level for many minutes and then retrace the same curve so long as the normal resonance curve is not met. Increasing the  $H_1$  field to 0.12 Oe, the largest field that could be used in this experiment, does not produce any more than two large jumps.

Figure 2 shows the value of the  $H_1$  field for which the onset of nonlinear behavior occurs as a function of temperature and static field. Recent theoretical work by Liu and Brinkman<sup>7</sup> suggests that the nonlinear phenomena observed in this work may be due to a variation over the sample volume of the direction of the rotation axis'  $\hat{\omega}$  of the B phase. It appears from their work that nonlinear phenomena can only occur when the rotation axis  $\hat{\omega}$  is not aligned with the static field  $\tilde{H}_0$ . The field and temperature dependence



FIG. 2. The  $H_1$  field for which the first nonlinear event is observed as a function of reduced temperature at a pressure of 21 bar. The solid curves are a fit to the data assuming  $H_1 = A + B/(1 - T/T_c)^{1/2}$ : O, 308 Oe,  $A = -0.0128 \text{ Oe}$ ,  $B = 0.0243 \text{ Oe}$ ;  $\bullet$ , 180 Oe,  $A = 0.0210$ Oe,  $B = 0.0121$  Oe;  $\Box$ , 102 Oe,  $A = -0.0021$  Oe,  $B =$ 0.0076 Oe, **m**, 31 Oe, data.

of the variation of  $\hat{\omega}$  in a right circular cylinder has been studied both theoretically and experimas been studied both theoretically and experi-<br>mentally by Brinkman  $et al.^9$  and arises from a competition between the field- and wall-orientation energies. The characteristic healing length is proportional to  $H_0^{-1}(1-T/T_c)^{1/2}$ . This suggests that the  $H_1$  field for which the first nonlinear event occurs might be estimated to be proportional to the inverse of the healing length and in particular would predict a square-root singularity as  $T$  approaches  $T_c$ . The solid curves drawn through the 308-, 180-, and 102-Oe data displayed in Fig. 2 are of the form  $H_1 = A + B/(1$  $-T/T_c$ <sup>1/2</sup> with the values of A and B being given in the figure caption. The onset data are consisin the figure caption. The onset data are consis<br>tent with a  $(1 - T/T_c)^{-1/2}$  temperature dependenc but do not firmly establish it. The exact dependence of the  $H_1$  field at onset on the  $H_0$  field cannot be determined from these data. The spinlattice relaxation time  $T<sub>1</sub>$  is different for all three fields. As would be expected from earlier work,<sup>10</sup>  $T_1$  is nearly proportional to the inverse of the gradient field, which is proportional to  $H_0$ , and it might need to be considered in the comparison of the onset data at different fields. At  $T/T_c$ =0.87 for example, the values of  $T_1$  for the 308-, 180-, and 102-Oe data was 0.24, 042, and 068 msec, respectively, with only a slight dependence on temperature below  $0.99T_c$ . The 31-Oe data displayed in Fig. 2 behaved differently from



FIG. 3. The magnitude of the large jumps normalized to the weak-coupling value of the magnetization of  ${}^{3}$ He-*B* as a function of reduced temperature at a pressure of 21 bar. The solid symbols are the highest-frequency jumps. Triagnels are 308-Oe data; circles are 180-Oe data; squares are 102-Oe data. The slopes of the dashed lines are exactly proportional to the inverse square of the static field.

the higher-field data. In the temperature range  $T/T_c = 0.86-0.93$  no cw NMR signal was ever observed. In the temperature range  $T/T_c = 0.93-$ 0.96, the onset of the nonlinear phenomena tended to depend upon the history of the  $H_1$  field, with the value of the  $H_1$  field necessary for the onset of nonlinear behavior being larger when increasing  $H_1$ , than when decreasing it. Above  $T/T_c = 0.96$ the onset data were reproducible and independent of the direction of approach. From the work of of the direction of approach. From the work of Osheroff  $et al.$ <sup>11</sup> it is likely that the observed behavior in 31 Oe is a result of an aligning of the rotation axis perpendicular to the sample chamber walls.

The cw NMR phenomena observed in this work can be grouped into two classes of nonlinear events according to the size of the jump in magnetization. The events with small jumps in magnetization exhibit little temperature dependence. There can be as many as four small jumps on a single sweep. Both the magnitude of the jumps and the frequencies at which they occur have a slight linear dependence on the  $H_1$  field. The magnitudes of these jumps can be as large as 4% of the  $B$ -phase magnetization, but at any fixed temperature there is at least a factor-of-2 difference between the magnitude of the large and small jumps and frequently a factor of 10 or more.



FIG. 4. The reduced frequency at which the jumps in magnetization occur normalized to the center frequency  $\omega_0$  as a function of reduced temperature at a pressure of 21 bar. The open symbols are for the large jumps and the closed symbols are for the first nonlinear event. Triangles are 308-Oe data; circles are 180-Oe data; squares are 102-Oe data. The slopes of dashed lines are exactly proportional to the inverse square of the static field.

The behavior of the events with two large jumps in magnetization is shown in Fig. 3, where the magnitude of each jump, normalized to the weakcoupling value of the  $B$ -phase magnetization, $\delta$  is plotted as a function of reduced temperature. The higher-frequency jump is shown as a solid symbol and the lower as an open symbol. The magnitude of the jumps is nearly independent of the  $H_1$  field and is temperature independent up to some temperature, which appears to depend only on  $H_0$ . Above this temperature the magnitude of the jump decreases linearly with increasing temperature initially, and then somewhat faster as  $T_c$  is approached. For each value of  $H_0$  there is a temperature above which no large jumps occur and only the nearly temperature-independent small jumps are observed. The slopes of the dashed lines drawn through the data are exactly proportional to the inverse of the square of the static field.

The frequency at which the large jumps occur, normalized to the center line frequency  $\omega_0$ , is shown as a function of temperature and field in Fig. 4. The two open symbols at each temperature represent the frequencies at which the two large jumps occur. These data are remarkably similar to those displayed in Fig. 3. The solid symbols are the frequencies at which the first

nonlinear event occurs. The slopes of the dashed lines drawn through the data are again exactly inversely proportional to the square of the  $H_0$  field. There is a slight dependence on the  $H_1$  field of the frequency at which the large jumps occur. The larger  $H_1$ , the higher the frequency at which the jump occurs.

The role that the gradient in magnetic field plays in these nonlinear phenomena cannot be unambiguously determined from the measurements presented here. However, an estimate of the importance of the gradient field may be made by comparing the experiments reported here with those of Ref. 1 in which the gradient was nearly 100 times smaller. Assuming that the nonlinear events depend primarily upon the magnitude of  $\Delta M_B/M_B$  as in the ferromagnetic resonance case, then the two experiments can be compared by using the standard relationship<sup>12</sup>  $\Delta M_B/M_B = \gamma^2 H_1^2$  $\times T_1T_2$ . I find, using the value for  $T_1$  and  $T_2$  appropriate for the two examples of Ref. 1 in 623 Oe at a reduced temperature  $T/T_c = 0.60$ , that no nonlinear phenomena were observed for  $\Delta M_B/M_B$ Nothing the formula were conserved for  $\Delta M_B / M_B$ <br> $\approx 5 \times 10^{-5}$  while for  $\Delta M_B / M_B$  $\approx 2 \times 10^{-2}$  the first nonlinear event was observed. In the present work, at the lowest temperature and highest field employed, the onset of nonlinear phenomena began at  $\Delta M_B / M_B = 1.1 \times 10^{-2}$ . Although this comparison is based on only two events, it does suggest that

the only major effect of the gradient field is to change the values of  $T_1$  and  $T_2$  and it does not play a dominant role in the physics of these interesting and unexplained nonlinear phenomena.

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# Ornstein-Zernike Theory of Classical Fluids at Low Density

D. B. Abraham<sup> $(a)$ </sup> and H. Kunz

Laboratoire de Physique Théorique, Ecole Polytechnique Fédérale, Lausanne, Switzerland (Received 26 April 1977)

We show rigorously that for dilute classical systems with finite-range interactions the pair-correlation function has the form predicted by Ornstein and Zernike.

The Ornstein-Zernike theory of pair-correlation functions is a cornerstone, albeit a heuristic one, in discussion of fluids and lattice gases outtion functions is a cornerstone, albeit a heurione, in discussion of fluids and lattice gases<br>side the critical region.<sup>1,2</sup> It predicts that the truncated pair-correlation function for a fluid  $u(\vec{r}, \rho)$  should behave asymptotically as

$$
u(\vec{\mathbf{r}}, \rho) = A \exp(-k_1 |\vec{\mathbf{r}}|) \cos k_2 |\vec{\mathbf{r}}|/|\vec{\mathbf{r}}|^{(d-1)/2}, \qquad (1)
$$

where  $A$ ,  $k_1$ , and  $k_2$  are functions of density and temperature and  $d$  is the dimensionality. In this Letter we establish the theory rigorously both for continuum systems with finite-range potentials and for lattice systems under the same con-

ditions, essentially for low densities and high temperatures. The latter extends results obtained by transfer-matrix techniques in this region to non-nearest-neighbor interactions in the transfer direction. Paes-Leme and Shor have obtained similar results for lattice gases independently. ' Prior to this work, bounds had been obtained on the spatial behavior of correlation functions.<sup>4</sup> Our results should prove useful in a field-theo $ext{set}$  results should prove useful in a freta-theorem retic context,<sup>5</sup> and more generally in statistical mechanics.<sup>6</sup>

The outline of our approach is as follows: We use the direct correlation function  $c(\vec{r}, \rho)$  first