<sup>4</sup>Sheldon Schultz and Gerald Dunifer, Phys. Rev. Letters 18, 283 (1967).

<sup>5</sup>F. J. Dyson, Phys. Rev. 98, 349 (1955).

<sup>6</sup>M. Lampe and P. M. Platzman, Phys. Rev. <u>150</u>, 340 (1966).

<sup>7</sup>In principle, an absolute measurement of the susceptibility determines the value of the isotropic exchange parameter. In practice, this measurement [R. Schumacher, T. Carver, and C. P. Slichter, Phys. Rev. <u>95</u>, 1089 (1954)] is extremely difficult to perform and gives results which necessarily have poor accuracy.

<sup>8</sup>P. Nozières, <u>Theory of Interacting Fermi Systems</u> (W. A. Benjamin, Inc., New York, 1964).

 $^{9}$ The mass  $m^{*}$  is the effective quasiparticle mass which includes band-structure effects, phonons, and

electron-electron effects. It can be shown {see M. Ya. Azbel', Zh. Eksperim. i Teor. Fiz. 34, 766 (1958) [translation: Soviet Phys.-JETP 7, 527 (1958)]} that, to a high degree of accuracy, the mass measured in Azbel'-Kaner cyclotron resonance {M. Ya. Azbel' and E. A. Kaner, Zh. Eksperim. i Teor. Fiz. 32, 896 (1956) [translation: Soviet Phys.-JETP 5, 730 (1957)]} is, in fact, the quasiparticle mass.

<sup>10</sup>The long-wavelength susceptibility, Eq. (4), depends only on  $B_0$  and  $B_1$ . The terms in  $k^4$  in  $\chi(k,\omega)$  will involve  $B_2$  as well. Experiments involving small wavelengths should give information about the higher moments of the Fermi-liquid function.

<sup>11</sup>T. M. Rice, Ann. Phys. (N.Y.) <u>30</u>, 100 (1965).
 <sup>12</sup>W. M. Walsh, Jr., L. W. Rupp, Jr., and P. H. Schmidt, Phys. Rev. <u>142</u>, 414 (1966).

## OBSERVATION OF SPIN WAVES IN SODIUM AND POTASSIUM\*

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We report the first observation of spin waves in sodium and potassium at low temperatures. Utilizing the theory of Platzman and Wolff, we are able to deduce the first two Legendre coefficients of the Landau correlation function for a Fermi liquid.

In the course of making precise g-value and linewidth measurements of the conduction-electron spin resonance (CESR) in sodium foils at 1.4°K, utilizing the selective transmission technique, we found significant departures from the predictions of previous theories. Further investigation showed that there was extra structure in the vicinity of the CESR. It is the purpose of this Letter to show that the extra structure that we have observed is proof of the existence of spin waves in the metal which are excited by the applied rf field. We have verified the detailed theory of Platzman and Wolff<sup>2</sup> (henceforth called P-W), and furthermore, we are able to extract from the data the relevant Landau Fermi-liquid theory parameters  $B_0$ and  $B_1$ . We wish to point out that these spinwave data appear as a primary effect in themselves and vanish in the limit of zero electron correlation. We believe that this direct observability, plus the fact that we are doing a resonance experiment, will allow the measurement of the relevant parameters with a significant precision.3 Since we have seen the spin-wave signals in both sodium and potassium, we feel the technique is generally applicable to all the alkali metals and in principle to all metals.4

The basic experimental arrangement is similar to that which we have used to discover the CESR in several new metals.<sup>5</sup> The major change has been the use of solid dielectric cavities. which help to insure the parallelism of the surfaces of the ductile alkali samples while also protecting them from any deteriorating atmosphere. In brief, the technique consists of placing the sample between a pair of X-band cavities tuned to the same frequency in the presence of a uniform dc field  $H_0$ . Microwave power is coupled into one cavity (transmitter), and a sensitive superheterodyne receiver is connected to the second cavity (receiver). If the leakage between the cavities has been sufficiently reduced (by experimental care), any power that appears in the receiver cavity has been transmitted through the sample.<sup>6</sup> In the vicinity of a magnetic resonance, electrons absorb power within a skin depth on the transmitter side of the sample and carry this information via their nonequilibrium transverse magnetization to the far side of the sample where they radiate power.7 As is shown in P-W, at selected values of the applied dc field near the main CESR (for a fixed frequency), other modes of the spin system are excited depending on

the properties of the interacting electron gas and the thickness of the sample. In Fig. 1 we have presented a set of typical data taken with the dc field parallel to the sample ( $\Delta = 90^{\circ}$ ). Under this condition the spin-wave structure appears on the low-field side of the main CESR signal. As the magnetic field is rotated towards the normal to the sample, the spin-wave structure moves in towards the main CESR line with all the modes converging at a particular angle  $\Delta_{\mathcal{C}}$ . When the field is rotated still further, the structure appears on the high-field side and is farthest out when the field is normal to the sample ( $\Delta = 0^{\circ}$ ). The magnitude of the separation of the spin waves from the main CESR line is found to be almost proportional to  $1/L^2$ , and the successive harmonics are separated proportional to  $n^2$  as is predicted by the theory (L is the sample thickness, nis the harmonic number). For a given thickness, the intensity and amplitudes of the spin waves are mainly determined by the electron collision time  $\tau$ , and as  $\tau$  decreases, the lines

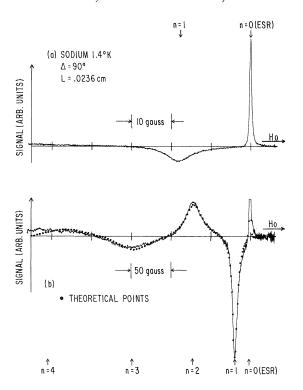


FIG. 1. Typical spin-wave signals as a function of applied dc magnetic field ( $H_0 \approx 3250$  G). (a) The n=0 mode (usual CESR) and the n=1 mode are clearly shown. (b) The gain and field sweep have been increased to display the first four spin-wave modes beyond the CESR. The theoretical points have been fitted via the procedure described in the text.

broaden, diminish in intensity, and shift slightly. For Na we have been able to follow these signals up to  $\approx 11^{\circ}$ K. It is of interest to note that the area under each of the first few spin waves is greater than that of the main CESR.

We have programmed the results of the full formula for the transmitted rf field developed by P-W [Eq. (5)], and in Fig. 1(b) the linewidth and location of the first spin wave have been fitted to the experimental data by adjusting  $\tau$ and  $B_0$ , respectively, and by normalizing the signal at the peak of the first spin wave. As can be seen, the agreement is generally excellent but falls off at the higher order spin waves. The mathematical reasons for this are understood, and it is very likely that a treatment to higher order in the expansion parameters of P-W will be necessary to obtain better agreement with experiment. It is thus possible that the higher order modes will allow us to obtain information about  $B_2$  or even higher order coefficients.

Equation (5) of P-W is of sufficient complexity that is is impractical to discuss the analysis of the data for arbitrary values of all of the parameters in this brief account. Let us impose the following two conditions: (1)  $\omega\tau$   $\gg$  1,8 and (2) the separation of the first spin wave maximum from the main CESR ( $\alpha_{\Delta}$ ) at any angle  $\Delta$  is sufficiently small. The first condition is necessary if one wishes to make precision measurements, and the second can always be met by taking sufficiently thick films. Under these assumptions the transmitted spin wave signal is given by [i.e., Eq. (5) of P-W reduces to]

$$H_t \cong [T_2 \Gamma^2 W \sin(2W)]^{-1},$$

with

$$\begin{split} -4W^2 &= (\alpha+i)/T_2\Gamma^2, \quad \alpha = (\omega-\omega_S)T_2, \\ \Gamma^2 &= \frac{v_F^2(B_0-B_1)(1+B_1)}{3L^2\omega_S} \\ &\times \left[\frac{\sin^2\!\Delta}{k^2(1+B_1)^2-\left[(B_0-B_1)/(1+B_0)\right]^2} \right. \\ &\left. -\frac{\cos^2\!\Delta}{\left[(B_0-B_1)/(1+B_0)\right]^2}\right], \end{split}$$

where  $T_{\rm 2}={\rm spin}$  relaxation time,  $\omega={\rm applied}$  frequency, k=m/m\*,  $v_{\rm F}={\rm Fermi}$  velocity, and  $\omega_{\rm S}={\rm CESR}$  frequency.

Quantitative analysis of the data. (A) Relative data. The basic term causing the resonances is  $1/\sin(2W)$  which blows up when  $\operatorname{Re}(2W) = n\pi$ . In the limiting case considered,  $T_2\Gamma^2$  is pure real (the neglected imaginary part determines the finite width and amplitude), and we can set  $n^2\pi^2T_2\Gamma^2 = -\alpha_\Delta$ . We can then express the ratio of the location of the spin wave at arbitrary  $\Delta$  to that at  $\Delta = 90^\circ$  by

$$\alpha_{\Delta}/\alpha_{90} = 1 - A \cos^2 \Delta \tag{1}$$

where  $A = k^2 (1 + B_0)^2 (1 + B_1)^2 / (B_0 - B_1)^2$ . If  $m^*$ is known, 9 we see that a measurement of the angle  $\Delta_C$  for which  $\alpha_{\Delta_C} = 0$  determines A and hence a relation between  $B_0$  and  $B_1$ . In Fig. 2 we show a plot of Eq. (1) normalized to the data at  $\Delta = 0^{\circ}$ . Using the observed value of 7.35 for the ratio at 0°, we obtain  $\Delta_C = 69.7 \pm 0.4$ °. Independently, we have measured the angle at which the spin waves coalesce and find the same  $\Delta_C$  to within ±0.5°. Notice that this is a fundamental property of sodium and does not depend on the sample thickness, etc. Figure 3 shows the relationship between  $B_0$  and  $B_1$  determined from this data. We believe the relationship between  $B_0$  and  $B_1$  shown in Fig. 3 is accurate to  $\pm 0.01$  in the region of interest.

(B) Absolute data. – In order to fix the abso-

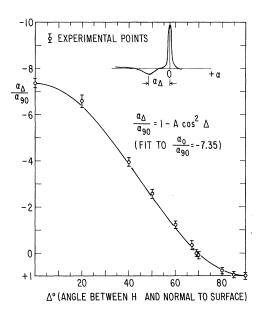


FIG. 2. The ratio of the location of the maxima of the first spin wave at arbitrary angle  $\Delta$  to the location at  $\Delta = 90^{\circ}$ , versus the magnet angle  $\Delta$ . The data were normalized at  $\Delta = 0^{\circ}$ . These data enable one to determine the parameter A.

lute value of  $B_0$  (and hence  $B_1$ ), it is necessary to fit the observed separations as a function of the other parameters in the theory. Subject to our two assumptions, and specifically considering  $\Delta=0^\circ$  for simplicity [other angles are simply related by Eq. (1)], we find

$$\alpha_0 = Q \left( 1 + B_0 \right) \tag{2}$$

where Q is a function of  $v_{\mathbf{F}}/L$ ,  $m^*$ , and A. (Note that  $T_2$  is not considered a parameter as it enters solely as a scaling factor in this expression.) Thus, since the linewidths of the spin waves are almost completely determined by  $\tau$  (for reasonable values of  $B_0$ ), we fit the data of Fig. 1 by adjusting  $\tau$  and then choosing the  $B_0$  which locates the maximum of the first resonance. Since  $B_0$  turns out to be small compared to  $1 \approx 10\%$ ), it is necessary to have 10 times the precision in  $\alpha/Q$  for a given precision of  $B_0$ . This imposes a rather severe limit to our present accuracy of the absolute values of  $B_0$ . Using our measured value of the thickness of the sample (corrected for thermal contraction) and a value of  $0.86 \times 10^8$  for  $v_{\rm F}$ , 11 we find  $B_0 = -0.1 \pm 0.1$ . From Fig. 3 this implies  $B_1 = 0.2 \pm 0.2$ . Although our present absolute accuracy is poor, we feel we can make an order-of-magnitude improvement (as discussed later) and hence we do not feel it worthwhile to elaborate on the errors in our current measurements.

There are other features related to the ob-

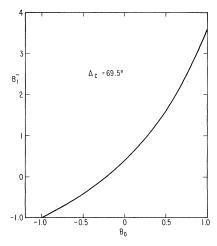


FIG. 3. The parameter  $B_1$  as a function of  $B_0$ . The equation relating them via the parameter A of Fig. 2 is described in the text.  $B_0$  and  $B_1$  are normalized Legendre coefficients of the spin part of the Landau correlation function. See the preceding paper (Ref. 2) for their precise definition.

servation of the spin waves that are worth mentioning: (1) "Geometric cyclotron resonance" effects. Under the conditions that  $\omega \tau > 1$  and sample thicknesses are comparable to the mean free path, one finds additional transmitted power variations with magnetic field angle and strength. We have previously termed these variations "geometric cyclotron resonances." Ordinarily the presence of these extra signals tends to obscure the spin waves and cause baseline shifts, but under a judicious choice of sample conditions they can be suppressed.

On the other hand, it is clear that in order to obtain the desired improvement in accuracy, it will be necessary to determine  $v_{\rm F}$  accurately. Since it is possible to determine  $k_{\rm F}$  accurately, or even more desirably,  $k_{\rm F}/L$ , we will always need  $m^*/m$  accurately. We have just completed an extensive study of these geometric resonances, and while a detailed theoretical description is still lacking, we feel that for materials with spherical Fermi surfaces we will be able to determine  $m^*/m$  to approximately 0.1% accuracy.

- (2) Determination of  $k_{\rm F}/L$ . If we can, in fact, measure the spin-wave data itself, and m\*/m to 0.1% accuracy, we will then need a precise determination of  $k_{\rm F}/L$ . We believe it would be possible to do Gantmakher size-effect resonance<sup>12</sup> and spin-wave resonance on the same samples at low temperatures. Since the size effect measures  $k_{\rm F}/L$  in terms of fundamental constants and the magnetic field at resonance, we should be able to achieve a substantial increase in accuracy. Thus, it may very likely be possible to make absolute determinations of the  $B_0, B_1$  (or even higher coefficients) with errors of a few percent.
- (3) The main CESR. So far, in the development of the P-W theory the main CESR does not play an intrinsic role in describing the spin waves, and  $T_2$  enters only as a phenomological spin relaxation time. However, we have observed striking changes in the main CESR (n = 0 spin wave) which indicates the need for further refinement in the theory. Due to the high precision with which the main CESR may be measured, this may be a sensitive probe of the interactions in the spin system. Briefly, we find significant changes in the g value, amplitude, and linewidth as the angle  $\Delta$  is varied, with the main variations occurring in a narrow region around  $\Delta_c$ . Over one order of magnitude changes in the linewidth have been observed

in our best samples. These line-shape changes are not due to a simple superposition of the spin waves coalescing at the main CESR because they do not show up in the P-W theory. Since this theory does take into account the changes in the "effective diffusion constant" that occur due to magnetic field effects, 13 this implies that other sources of explanation must be found. It is possible that it is merely a coincidence that these effects occur at  $\Delta_C$ , and they may be manifestations of changes in the rf field distributions in the bulk of the sample as were indicated in the experiment of Peercy and Walsh,14 or due to transit-time effects since the mean free paths are comparable to the thickness of the sample. All of the above variations gradually disappear as the sample temperature is raised, and are gone by ≈20°K. We plan to make an extensive study of these effects using pulse techniques, which are particularly suited to the transmission technique. We have made preliminary measurements of  $T_2^*$  (the freeinduction decay time) and it is in excellent agreement with the observed linewidths. 15 We now plan to measure  $T_2$  and  $T_1$  by the appropriate pulse techniques.

We wish to thank Professor D. Fredkin and Dr. P. Platzman for enlightening discussions on many-body effects, Mr. C. E. Taylor for generously supplying us with the pure sodium, and Mr. M. R. Shanabarger for his help in making the pulse measurements.

<sup>\*</sup>Work supported by the National Science Foundation. <sup>1</sup>M. Lampe and P. M. Platzman, Phys. Rev. <u>150</u>, 340 (1966), and our own detailed calculations (to be published).

 $<sup>^2\</sup>mathrm{P.}$  M. Platzman and P. A. Wolff, preceding Letter [Phys. Rev. Letters <u>18</u>, 280 (1967)]. We use the notation of that work throughout. There the normalization of  $\boldsymbol{B}_0$  and  $\boldsymbol{B}_1$ , which are Legendre coefficients of the spin part of the Landau correlation function, is explicitly defined.

 $<sup>^3\</sup>mathrm{We}$  know of only one other technique for obtaining  $B_0$ , namely, a measurement of the absolute electronic susceptibility. This method has only been applied to Li and Na [R. Schumacher and C. P. Slichter, Phys. Rev. 101, 58 (1956)] and is not generally applicable to other metals for experimental reasons. Due to the nature of the experiments they are expected to have substantial errors. We know of no other technique for measuring  $B_1$  in metals.

<sup>&</sup>lt;sup>4</sup>Although we have seen spin waves in potassium qualitatively similar in behavior to those in sodium, we have not completed our analysis of those data. All the data in this Letter refer specifically to sodium. Since

we have now seen CESR in four metals besides all the alkalis, there is a good possibility for observing spin waves in at least one of them.

<sup>5</sup>S. Schultz and C. Latham, Phys. Rev. Letters <u>12</u>, 695 (1965); S. Schultz and M. R. Shanabarger, Phys. Rev. Letters <u>16</u>, 187 (1966).

 $^6$ This is a big "if." In our experiments leakage was typically better than 165 dB, and the actual power in the main <u>spin-wave peak</u> was ≈20 dB <u>above leakage</u> for the data presented in Fig. 1.

 $^7$ Although we have detected the power transmitted, for most purposes it is more convenient to measure the component of the transmitted magnetic field projected on a reference rf field, and all the data shown were taken with the reference field adjusted so as to observe the imaginary part of the complex susceptibility (i.e., the quantity which is called  $\chi''$  in the usual resonance terminology.)

 $^8$ For our material  $ho(R.T.)/
ho(4.2^\circ K) \approx 6000$ , and the appropriate  $\omega \tau \approx 20$ . Making corrections due to the finite  $\omega \tau$  does not significantly alter any of the results presented.  $T_2 \approx 10^{-6}$  sec.

 $^9$ We have used the value of 1.24 for  $m^*/m$  in sodium. C. C. Grimes and A. F. Kip, Phys. Rev. <u>132</u>, 1991 (1963).

 $^{10}\mathrm{There}$  are, of course, two roots for  $B_1$  in the solution of the equation. One of these is eliminated in that it predicts the location of the spin waves on the wrong side of the CESR.

 $^{11}$ M. T. Taylor, Phys. Rev.  $\underline{137}$ , A1145 (1965), and using  $m^*/m = 1.24$ .

<sup>12</sup>V. F. Gantmakher, Zh. Eksperim. i Teor. Fiz. <u>42</u>, 1416 (1962) [translation: Soviet Phys.—JETP <u>15</u>, 982 (1962)].

<sup>13</sup>See Ref. 1 and G. D. Gaspari, Phys. Rev. <u>151</u>, 215 (1966)

<sup>14</sup>P. S. Peercy and W. M. Walsh, Jr., Phys. Rev. Letters 17, 741 (1966).

 $^{15}$ For the range of sample thicknesses considered, the diffusion time for the spin information is much less than the spin relaxation time. Hence, a free-induction decay measures  $T_2^*$ . The same equipment is being used for experiments at higher temperatures where the reverse relationship between the times applies.

## MEASUREMENT OF 2e/h USING THE ac JOSEPHSON EFFECT AND ITS IMPLICATIONS FOR QUANTUM ELECTRODYNAMICS\*

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Using the ac Josephson effect, we have determined that  $2e/h = 483.5912 \pm 0.0030 \text{ MHz}/\mu\text{V}$ . The implications of this measurement for quantum electrodynamics are discussed as well as its effect on our knowledge of the fundamental physical constants.

In this Letter, we report a high-accuracy measurement of 2e/h using the ac Josephson effect (here, e is the electron charge and h is Planck's constant). When combined with the measured values of other fundamental constants, this measurement yields a new value for the fine-structure constant  $\alpha$  which differs by 21 ppm from the presently accepted value. This change in  $\alpha$  removes the present discrepancy between the theoretical and experimental values of the hyperfine splitting in the ground state of atomic hydrogen, one of the major un-

solved problems of quantum electrodynamics today. We also discuss the effect of this change on our present knowledge of the fundamental physical constants.

The phenomenon used in these experiments was first predicted by Josephson in 1962. He showed theoretically that when two weakly coupled superconductors are maintained at a potential difference V, an ac supercurrent of frequency

$$\nu = (2e/h)V \tag{1}$$