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Spin waves in lithium at 80 GHz

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The use of a microwave transmission spectrometer operating at 80 GHz has enabled the first observation of resolved spin waves in lithium. Comparison of the experimental data with theoretical line shapes derived from the Landau Fermi-liquid theory allows a determination of the Landau many-body spin parameters B_0 and B_1 for a series of effective-mass (m^*) values between $2.1m_0$ and $2.5m_0$ where m_0 is the free-electron mass. By comparing with data on sodium, potassium, and rubidium we deduce that the most likely values for these three parameters are $m^*/m_0=2.30\pm0.20$, and for a mass ratio of 2.30 that $B_0=-0.23\pm0.03$ and $B_1=-0.08\pm0.05$. Moreover we find that the ratio of the Pauli spin susceptibility to that of a free-electron gas is 2.99 ± 0.12 irrespective of m^*. All these values compare favorably with other measurements.

INTRODUCTION

Although spin waves have been resolved in sodium,[1,2] potassium,[1,2] and rubidium,[3] no such observations have yet been made in lithium. This is primarily due to the relatively low-purity and consequentially low-momentum scattering time (τ) of the available material. However, observations of asymmetries in the line shape of the conduction-electron spin resonance (CESR), as a result of unresolved spin waves, have been recorded by Flesner and Schultz[4] using a microwave transmission spectrometer operating at 9.5 GHz. Because the strength of the spin-wave modes depends, in part, upon $\omega\tau$, where ω is the microwave frequency, using a higher frequency (80 GHz) has enabled the first observation of resolved spin waves in similarly low residual-resistivity-ratio (RRR) lithium. Subsequent determination of the B_n parameters characterizing the spin part of the many-body electron-electron interaction requires the additional knowledge of the effective mass (m^*) for lithium. However, unlike the other alkali metals, m^* is not precisely known for lithium, therefore it is not possible to uniquely analyze the data. Instead we present several pairs of possible values for the B_0,B_1 parameters where each pair is determined assuming a different m^* lying in the range $2.1m_0$ to $2.5m_0$, m_0 being the free electron mass. We also obtain spin susceptibility values for this range of m^* and find the ratio of the Pauli spin susceptibility χ to that of a free-electron gas having the same density, χ_0, is constant irrespective of m^*.

THEORY

The spin-wave data previously obtained in the alkali metals have been analyzed using two theories, that of Platzman and Wolff[5] (to be referred to as PW) and that of Wilson and Fredkin[6] (to be referred to as WF), both of which have been derived from the Landau Fermi-liquid theory. While the PW theory is a good approximation for low wave vectors, the WF theory is exact, incorporating any number of B_n parameters. In addition, WF treat the relaxation times τ and T_2 (spin relaxation time) more exactly. Using the WF algorithm in the form of computed line shapes following closely the programs developed by Dunifer *et al.*,[1] we are able to fit the experimental data and thereby obtain the relevant spin-wave parameters. In this fitting procedure one needs to know m^*/m_0 the effective-mass ratio, V_F the Fermi velocity, g the Landé g factor, the sample thickness, and the frequency of the incident microwave radiation. While the latter three parameters may be readily obtained, it is not possible to define m^*/m_0 and V_F uniquely. Hence the fitting procedure has been carried out over several values of m^* keeping the product m^*V_F constant.

For a particular m^* it is found that for any given sample (for which the magnetic field is held at a constant orientation relative to the surface) there are a large number of combinations of B_0 and B_1 which will give good line shape fits with appropriate adjustments of τ and T_2. It is necessary to have two different orientations of the field in order to uniquely determine B_0 and B_1 separately. Our samples are oriented with the applied field either perpendicular or parallel to the surfaces. One may use the PW theory to show that to a first approximation, the parameters $f_\perp$ and $f_\parallel$ defined through

$$f_\perp=\frac{(1+B_1)(1+B_0)^2}{B_0-B_1}$$

and

$$f_\parallel=\frac{(1+B_1')(1+B_0')^2}{(B_0'-B_1')}\left[\left[\frac{(1+B_0')(1+B_1')}{\alpha(B_0'-B_1')}\right]^2-1\right]^{-1},$$

where $\alpha=gm^*/2m_0$ are the parameters which primarily define the spin waves for the perpendicular and parallel geometries, respectively. Approximate values for these parameters are obtained for the two orientations from the first attempts at fitting the data. Then one may deduce better values for B_0 and B_1 by inverting the two equations

$$B_0=\frac{-f_\perp}{\alpha\left[1+\frac{f_\perp}{f_\parallel}\right]^{1/2}}-1 \tag{1}$$

and

$$B_1=\frac{(1+B_0)f_\perp}{(1+B_0)^2+f_\perp}-1\;. \tag{2}$$

These are then used to produce a new fit to the experimental spin-wave line shape. Suitable small adjustments of τ, T_2, and B_0 are then made to give the best fits and new $f_\perp, f_\parallel$ values obtained. The process is then repeated but this time making small adjustments to B_1 instead of B_0. At this point the numbers for B_0 and B_1 converge, to within suitable error limits, to the same values for all samples and it is these values which are chosen as the best values for these parameters.

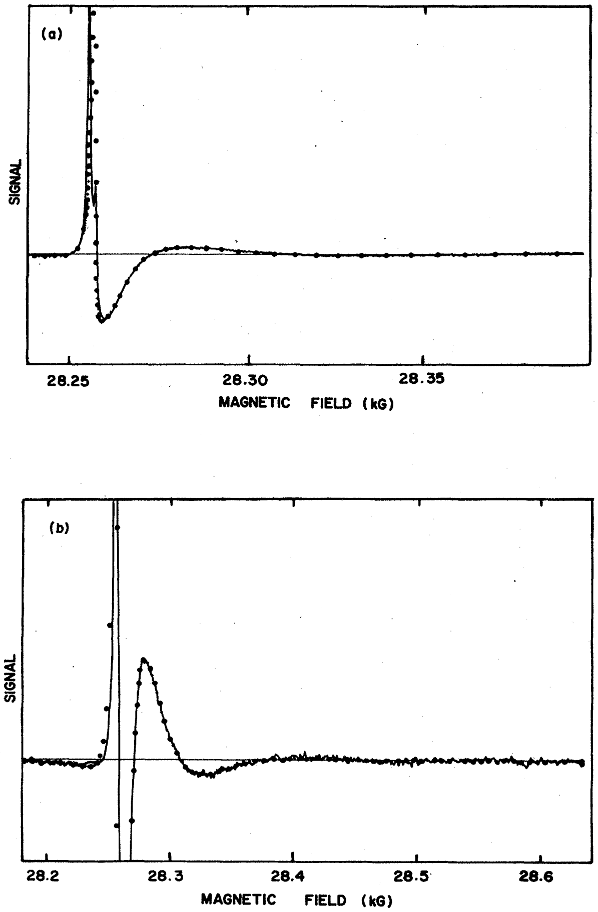

FIG. 1. The spin-wave line shape (continuous line) for a 165-μm slice observed at 79.19 GHz and 1.4 K in the field-perpendicular configuration is compared with that produced theoretically (circles) using the WF theory and $m^*/m_0=2.3$. (a) Narrow sweep, using $T_2=0.95\times10^{-7}$ s, $\tau=0.72\times10^{-11}$ s, $B_0=-0.225$, and $B_1=-0.060$ in the theory. (b) Large sweep with the gain ~10 times greater than in (a) using $T_2=1.10\times10^{-7}$ s, $\tau=0.81\times10^{-11}$ s, $B_0=-0.225$, and $B_1=-0.075$ in the theory.

EXPERIMENTAL TECHNIQUE

The transmission spectrometer used to observe the spin waves has been described briefly in an earlier paper by Mace *et al.*[7] and in more detail by Dunifer and Pattison,[8] while details of the resonant cavities are given by Mace *et al.*[2] The samples were all prepared in an argon-filled glove box in which the H_2O, O_2, and N_2 levels were less than 1 ppm. For samples thicker than 150 μm the lithium was initially extruded as a ribbon, whereas for thinner samples it had to be squashed between two pieces of lightly greased glass in order to achieve a thickness $\sim10\%$ greater than the final desired thickness. The subsequent compression of the material between the two cavities at 800 psi for 5 min took place in a vacuum chamber at room temperature to avoid the trapping of argon gas between the lithium and the quartz windows on each side of the sample. The samples were then cooled as quickly as possible to liquid-helium temperatures to minimize the amount of martensitic phase transformation. Typically they were cooled from room temperature to 100 K in ~20 min and then to 4.2 K in ~8 min. The lithium supplied by the Foote Mineral Company had a bulk RRR of 1000 ± 200.

RESULTS

In both Figs. 1 and 2 we show spin-wave data for lithium together with our final line-shape fits using the WF theory. Figures 1(a) and (b) are spin waves observed in a 165-μm slice with the field oriented perpendicular to the sample, while Figs. 2(a) and (b) are those observed in a 112-μm slice with the field oriented parallel to the sample. The apparent splitting of the CESR in Fig. 1(a) is due to significant magnetic field inhomogeneity over the sample in the field-perpendicular configuration. This is substantially reduced in the field-parallel arrangement as is clear in Fig. 2(a). The inhomogeneity for the field-perpendicular geometry corresponds to a variation in field of about 5 parts in 10^5 across the sample which is 4 mm long. This exceeds the specifications for the 40-mm-diam-bore superconducting solenoid used to generate the field, and the source of the inhomogeneity is presently unknown. Because the same effects occur in other metals observed in this geometry, the effect is clearly not associated with the lithium metal per se.

As seen in the figures, the spin waves are clearly very weak even at 80 GHz so it is not surprising that they have been unresolved in previous experiments at much lower frequencies. In none of the samples was it possible to observe any geometric resonances associated with the orbital motion of the electrons in the magnetic field.

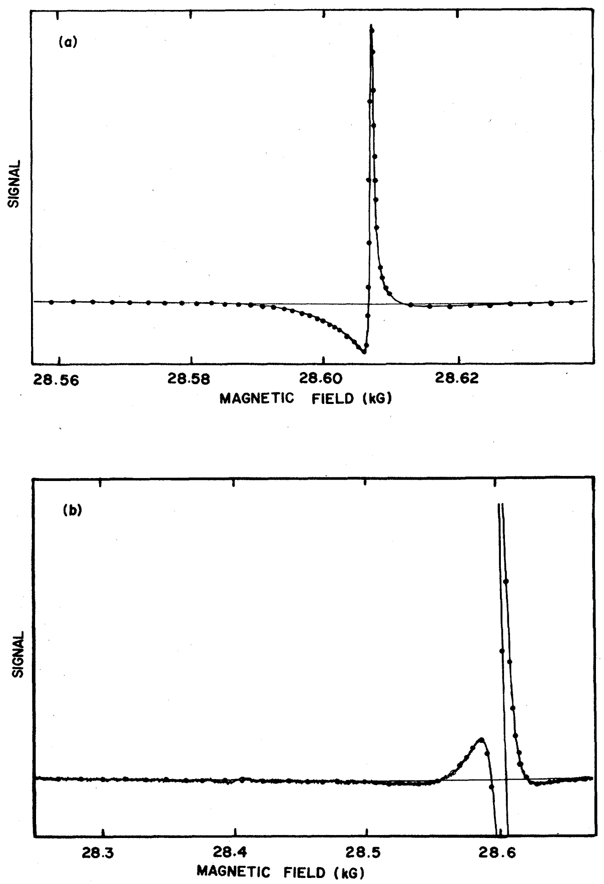

FIG. 2. The spin-wave line shape (continuous line) for a 112-μm slice observed at 80.17 GHz and 1.4 K in the field-parallel-to-sample-surface configuration is compared with that produced theoretically (circles) using the WF theory and $m^*/m_0=2.3$. (a) Narrow sweep, using $T_2=1.60\times10^{-7}$ s, $\tau=0.95\times10^{-11}$ s, $B_0=-0.225$, and $B_1=-0.080$ in the theory. (b) Large sweep with the gain 10 times greater than in (a) using $T_2=1.45\times10^{-7}$ s, $\tau=0.92\times10^{-11}$ s, $B_0=-0.225$, and $B_1=-0.063$ in the theory.

DISCUSSION

In applying the WF theory we have presumed that $B_2=0$. In view of the lack of observation of high wave-vector spin waves this assumption is of no significance. Furthermore, data on both sodium and potassium suggest B_2 is small ($|B_2|<0.02$) for both metals,[2] and its influence may also be reasonably neglected for lithium. Hence we have values for B_0 and B_1 using the procedure outlined above, for given m^* values. There is unfortunately a serious problem in deciding the most appropriate m^* value for lithium. As a result of a partial martensitic phase transformation[9,10] from a body-centered-cubic structure to a hexagonal-close-packed structure, as the lithium is cooled below about 78 K, de Haas—van Alphen measurements to determine m^* in single-crystal lithium have so far not been possible. However, Randles and Springford[11] have observed some weak oscillations in samples of lithium particles dispersed in paraffin wax. From these oscillations they give a value for the low-temperature cyclotron effective mass of lithium of $(1.8\pm0.4)m_0$. However, the most accurate determinations of m^* that include electron-phonon effects are from specific heat measurements.

Filby and Martin[12] measured several different isotopic compositions of 99.9% pure lithium and their results yield an m^* of $2.20\pm0.02m_0$ with no indication of the influence of the martensitic phase transition upon their data. (The same author's data give $m^*=1.26\pm0.02m_0$ for sodium in excellent agreement with the de Haas—van Alphen value of 1.256 ± 0.003 of Elliott and Datars.[13]) Therefore, with no geometrical resonance signals observed in our experiment from which m^* may be deduced, this value of m^* is chosen as a starting point from which to fit the spin-wave data.

The lack of signals from the orbital motion of the electrons, e.g., the Gantmakher-Kaner oscillations, cyclotron phase resonance, and cyclotron waves,[14] may be partially attributed to the martensitic phase transformation. However, according to Schmidt[15] it is more probable that calcium impurities are responsible for the high scattering rate (10^{11} Hz). Because of the lack of geometric resonances not only m^* but also the sample thickness cannot be specified as for sodium and potassium. Consequently the only thickness determination comes from direct measurement using a precision micrometer. This value, after being corrected by 2.3% for thermal contraction[16] was accurate to ±3 μm. Additional parameters used in the calculations were $g=2.0023$ and $V_F=1.294\times10^6$ (m_0/m^*) m s^{-1}, the latter calculated from the measured lattice constant[10] assuming one conduction electron per atom.

We see in Figs. 1 and 2 very good agreement between the theory and the data, although as already mentioned field inhomogeneity in the field-perpendicular arrangement creates some discrepancy around the CESR. These field inhomogeneities are also responsible for the slight differences found between the relaxation times used for the fits to the low- and high-gain data. The agreement of the B_0 and B_1 values used for the line-shape fits for each sample has been forced by our fitting procedure as indicated above and the overall errors in B_0 and B_1 are substantially larger than the discrepancies observed between the samples. Details of the samples analyzed with their corresponding thickness errors are listed in Table I. Using the convergence method discussed above yields B_0, B_1 values from comparisons of both samples 1 and 2, and samples 1 and 3. The two sets of results for each m^* are in very close accord and yield the mean values shown as a

TABLE I. Samples used in the determination of the Fermi-liquid parameters.

Sample	Thickness at 4.2 K (μm)	Frequency (GHz)
1, field perpendicular	165 ± 3	79.2
2, field parallel	160 ± 3	79.8
3, field parallel	112 ± 3	80.2

function of m^* in Fig. 3. In this iterative fitting procedure it was found that B_0 could be shifted by ±0.02 before the B_1 value needed to fit the field-perpendicular data was no longer within the range of B_1 values required for the field-parallel data. This "fitting error" in B_0 is the dominant source of uncertainty in our measurements. The dependence of B_0 on sample thickness is considerably less for our present values of $\omega\tau$ than in the high-$\omega\tau$ limit. For example, we find under present conditions of analysis that a shift of 2% in sample thickness leads to a shift in B_0 of less than 0.01. The combination of these two errors leads to our quoted uncertainties of ± 0.03 in B_0 and consequently ± 0.05 in B_1.

Examination of the literature reveals only one other direct experimental determination of B_0 for lithium. Flesner and Schultz[4] analyzed line-shape asymmetries in the CESR of lithium observed at 9.5 GHz to obtain a B_0 of -0.23 ± 0.03 for an m^* of $2.21m_0$, assuming $B_1=0$. These numbers yield a somewhat lower $f_\perp$ value of -2.6 ± 0.5 which is within the error of our value of -3.3 ± 0.6 at $m^*=2.2m_0$ obtained from $B_0=-0.27\pm0.03$ and $B_1=-0.13\pm0.05$.

Further evidence to allow us to put limits on B_0 and B_1 and hence m^* comes from the data on sodium, potassium, and rubidium. These alkali metals have B_0 values of -0.22, -0.29, and -0.21 and B_1 values of -0.03, -0.07, and $+0.03$ respectively. This suggests that B_0 is likely to be of order -0.15 to -0.35 and B_1 is probably in the range -0.15 to $+0.05$. The B_0 range effectively encompasses our m^* range shown in Fig. 3, although the range in B_1 does suggest a somewhat higher m^*, in the region $2.15m_0$ to $2.65m_0$. We are led then to suggest that the most plausible value for m^* is $(2.3\pm0.2)m_0$ with corresponding B_0 and B_1 values for $m^*=2.3m_0$, of -0.23 ± 0.03 and -0.08 ± 0.05, respectively.

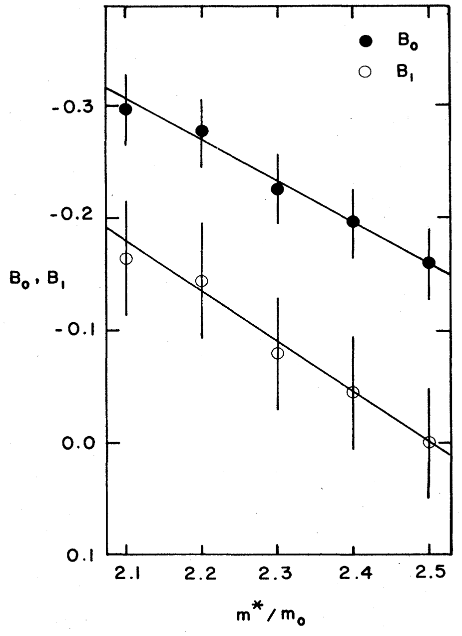

FIG. 3. Graphs of B_0 and B_1 versus m^*/m_0.

There is one other parameter which may be deduced from the data, that is the Pauli spin susceptibility χ, which in the Landau Fermi-liquid theory is given by

$$\frac{\chi}{\chi_0}=\frac{(g)^2}{(g_0)^2}\frac{m^*}{m_0}\frac{1}{1+B_0},$$

where the g value of lithium may be taken equal to the free-electron value g_0 to the accuracy necessary for interpreting the present experiment. Using the B_0's shown in Fig. 3, we find that

$$\frac{\chi}{\chi_0}=2.99\pm0.12,$$

irrespective of the choice of m^*. In fact, the individual χ/χ_0 values agree to within ±0.02 which is quite remarkable and suggests that this parameter is an invariant of the line shape fitting. Why this should be the case is not known. Within the framework of the WF theory the parameters B_0, B_1, and m^* are all independent phenomenological parameters, and there is no obvious constraint among them which would require χ/χ_0 to remain constant as they are varied during the line-shape-fitting process. As a consequence of this, however, our number may be compared in an absolute fashion with other determinations of χ/χ_0 for lithium. Very recently Vier *et al.*,[16] using transmission CESR in bilayers, have determined a value for this parameter of 2.86 ± 0.16 in good agreement with our value, while the earlier study by Flesner and Schultz[4] on damped spin waves gave 2.84 ± 0.1 at $m^*=2.21m_0$ and 2.93 ± 0.1 at $m^*=2.34m_0$. In Table II we summarize the primary experimental determinations for χ/χ_0 in lithium. We also note that the most recent theoretical result[17,18] for χ/χ_0 of 2.91 is in excellent agreement with our measured value. It is apparent from the table that values of χ/χ_0 determined from the spin waves and the integrated intensity of the CESR signal differ from each other by more than the sum of the quoted uncertainties. It is interesting that this is also the case

TABLE II. Experimental values for χ/χ_0 for indicated m^*/m_0 values. Values are listed for a temperature of 5 K. Any room temperature determinations have been scaled where necessary by assuming an 0.8% bulk thermal contraction between room temperature and 5 K (Ref. 16).

Type of observation	χ/χ_0	m^*/m_0
Present data on resolved spin waves	2.99±0.12	2.1–2.5
Transmission CESR from bilayers[a]	2.86±0.16	
Partially damped spin waves[b]	2.84±0.1	2.21
	2.93±0.1	2.34
Completely damped spin waves[b]	3.13±0.3	
Transmission CESR amplitude[c]	2.86±0.7	
Integrated intensity[d]	2.72±0.04	
Integrated intensity[e]	2.72±0.05	

[a] Reference 16.
[b] Reference 4.
[c] Reference 19.
[d] Reference 20.
[e] Reference 21.

for sodium, the only other metal in which χ/χ_0 has been measured using both techniques. In lithium the spin-wave value is larger than the integrated-intensity value by about twice the sum of the uncertainties, while in sodium the spin-wave value is smaller, again by about twice the uncertainties. This discrepancy, although small, may be significant and could be an indication that the two experimental methods measure somewhat different quantities. A careful examination of the theoretical foundations of both types of measurements might be useful.

Finally it must be emphasized that only a limited number of samples have so far been studied. Further investigations will be made once the lithium has been repurified using the technique of high-vacuum fractional distillation.[15] A residual resistivity ratio of ~4000 should be achievable which may enable the orbital signals to be resolved as well as increasing the resolution of the spin waves. This should lead to precise m^*, B_0, and B_1 values.

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