# Measurement of the Pauli susceptibility of lithium and sodium by spin-resonance transmission

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(Received 21 October 1977)

The intensity of the conduction-electron spin-resonance transmission signal in the regime of completely damped spin waves has been measured as a function of specimen thickness in lithium at 300 K and in sodium at 77 K. Theoretical expressions for on-resonance intensity as a function of specimen thickness are characterized by a decay length  $\delta_s^* = (2D^*T_2^*)^{1/2}$ , where  $D^*$  is the magnetization diffusion coefficient and  $T_2^*$  is the transverse relaxation time. A fit of these expressions to the data gives values of  $\delta_s^* = 23 \pm 2 \ \mu m$  for lithium and  $44 \pm 5 \ \mu m$  for sodium at these temperatures. Measured values of  $T_2^*$  were  $(1.7 \pm 0.2) \times 10^{-7}$  sec for lithium and  $(3.7 \pm 0.5) \times 10^{-8}$  sec for sodium. Analysis of these measurements in terms of the Landau theory of Fermi liquids leads to a determination of the many-body enhanced susceptibility  $\chi/\chi^0$ , whose values are  $2.86 \pm 0.7$  for lithium and  $2.45 \pm 0.7$  for sodium. The result for lithium is in good agreement with other measurements, but the value for sodium is not. Surface relaxation effects appear to be important in sodium, and their contribution is discussed.

#### I.- INTRODUCTION

It has long been recognized that a satisfactory description of many-body interactions in metals must include both electron-phonon and electronelectron contributions. The electron-electron contribution to the properties of simple metals is often described in terms of the Landau theory of Fermi liquids, first proposed<sup>1</sup> for liquid <sup>3</sup>He and since suitably modified for charged Fermi systems.<sup>2</sup> In that theory the quasiparticle interaction function is assumed to be separable into spindependent and spin-independent parts. If the spin dependence is further assumed to originate in exchange forces, the spin-dependent term is of the form  $f^{a}(\mathbf{\bar{k}},\mathbf{\bar{k}}')\mathbf{\bar{\sigma}}\cdot\mathbf{\bar{\sigma}}'$ , where  $f^{a}(\mathbf{\bar{k}},\mathbf{\bar{k}}')$  is an antisymmetric function of  $\vec{k}$  and  $\vec{k}'$ , the wave vectors of the interacting quasiparticles. For a spherical Fermi surface,  $f^{a}(\mathbf{k}, \mathbf{k}')$  is a function only of  $\theta$ , the angle between k and k', and can be expanded in Legendre polynomials of  $\cos\theta$  with expansion coefficients conventionally designated  $B_n$ . When the theory is used to derive expressions for measurable quantities,<sup>3</sup> the deviations from the predictions of freeelectron theory are expressed in terms of the expansion coefficients  $B_n$  and other quantities that include electron-phonon and band-structure effects.

The conduction-electron dc paramagnetic susceptibility, for example, is found to be enhanced over the free-electron value  $\chi^0$  according to

$$\chi/\chi^{0} = (m^{*}/m)/(1 + B_{0}),$$
 (1)

where both  $m^*$  and  $B_0$  must include electron-phonon contributions. Although there have been some attempts to calculate numerical values of  $B_n$  directly from first principles,<sup>3</sup> most calculations do not take this approach, but rather attempt to calculate directly quantities such as  $\chi/\chi^0$ . The best test of these calculations is to compare them with values of  $\chi/\chi^0$  measured directly,<sup>4,5</sup> but such measurements have been possible only for lithium and sodium. One of us has recently reported improved direct measurements of  $\chi/\chi^0$  in these metals, along with a theory of the measurement that establishes its validity on very general grounds.<sup>6</sup>

Because the direct measurement of  $\chi/\chi^0$  is limited in its applicability, the problem has been attacked differently by others who, recognizing that values of  $m^*/m$  are available from measurements of cyclotron resonance. the electronic specific heat, or the de Haas-van Alphen effect, have carried out experiments to measure  $B_0$ . These experiments have been of two types. Schultz and collaborators have for several years carried out conduction electron-spin-resonance transmission experiments in the regime of resolved spin waves,<sup>7</sup> under conditions in which the spin waves are present but unresolved,<sup>8</sup> and under conditions in which the spin waves are completely damped,<sup>8</sup> and have determined  $B_0$  in most of the alkali metals. In another approach, de Haas-van Alphen measurements have been used to determine  $B_0$  in several alkalis, but not in lithium.9,10

The experiments to be described here utilize spin-resonance transmission, although they differ from those of Schultz. They are not recent, having been carried out several years ago,<sup>11</sup> motivated at that time by a large discrepancy between the directly determined value of  $\chi/\chi^0$  in sodium and the value then inferred from spin-wave measurements of  $B_0$ . They were the first attempt to determine  $\chi/\chi^0$  from spin-resonance transmission under conditions of completely damped spin waves. As originally conceived, they were intended to be measurements of  $B_0$ . Viewed that way, they

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seemed to share one property with the spin-wave measurements<sup>7</sup>: that the extraction of  $B_0$  from the experimental data required the assumption (or simultaneous measurement) of a value for  $m^*/m$ , and that the calculation of an enhanced susceptibility from Eq. (1) required that  $m^*/m$  enter again in a way that did not necessarily cancel. Thus the value of  $\chi/\chi^0$  inferred from measurements of  $B_0$  could depend significantly on the value of  $m^*/m$ .

Recent analysis of Fermi liquid theory has shown,<sup>12</sup> however, that measurements such as ours can yield the susceptibility enhancement without needing a value for  $m^*/m$ , and we prefer now to interpret our results as a measurement of  $\chi/\chi^0$ rather than of  $B_0$ . Although our initial interest was directed primarily to sodium, we also made measurements in lithium for which, until the recent work of Flesner and Schultz,8 no other determinations of  $\chi/\chi^0$  had been made except for the direct measurements. Our results in lithium are essentially in agreement with those of Flesner and Schultz, although not as precise as theirs. In sodium our results disagree with all other measurements and are, therefore, suspect. In describing these measurements we shall suggest possible reasons for the difficulty. Analysis of the sodium results required the inclusion of surface relaxation effects, and we shall describe how surface relaxation may be included in calculations of the transmission line shape.

Spin-resonance transmission may be observed in simple metals whenever the diffusion length for the magnetization is appreciably larger than the classical skin depth. In resonance transmission experiments a flat-plate specimen, thick compared with the skin depth, is excited at one surface by microwave radiation of frequency  $\omega_0$ . As the external magnetic field is varied through the resonance value  $H_0 = \omega_0 / \gamma$ , power is absorbed by the spins, resulting in a nonequilibrium transverse magnetization. As the electrons diffuse away from the surface, the magnetization continues to precess coherently for a transverse relaxation time  $T_2^*$ , during which it may be transported by diffusion to the far surface. At the far surface, the power radiated by the coherently precessing magnetization is detected, and the phase and amplitude are observed as a function of magnetic field. The amplitude of the transmitted signal varies with specimen thickness, with a characteristic attenuation length  $\delta_s^* = (2D^*T_2^*)^{1/2}$ , where  $D^*$  is the diffusion coefficient for the magnetization. For noninteracting electrons  $D^*$  will be identical with D $=\frac{1}{3}V_{\rm F}^2\tau$ , the diffusion coefficient for charge or mass transport; for interacting electrons  $D^*$  is no longer equal to D but is modified by the spindependent electron-electron interaction. Specifically,  $D^*/D$  depends on the Landau parameters  $B_0$  and  $B_1$ , but, since  $B_1$  is often much less than  $B_0$ ,  $D^*/D$  may depend essentially only on  $B_0$ .<sup>13</sup>

The experiments described here determine the attenuation length  $\delta_s^*$  from measurements of the transmitted intensity as a function of specimen thickness, and the relaxation time  $T_2^*$  from measurements of the line shape. These are combined to give a value for  $D^*$ . Comparison of  $D^*$  with values of D inferred from conductivity measurements, together with the predictions of Fermi liquid theory, then gives a value for the susceptibility enhancement  $\chi/\chi^0$ . These experiments share one essential feature with the spin-wave measurements, the de Haas-van Alphen measurements. and the recent transmission resonance measurements of Flesner and Schultz<sup>8</sup>: that a determination of  $\chi/\chi^0$  from the data relies crucially on the results of Fermi liquid theory. By contrast, measurements of  $\chi$  by the method proposed originally by Slichter and first carried out by Schumacher<sup>4</sup> are remarkably free from reliance on phenomenological theories.

## **II. THEORETICAL FOUNDATION OF THE MEASUREMENT**

#### A. Noninteracting electrons

Theoretical line shapes for resonance transmission by noninteracting conduction electrons can be described using the diffusion-modified Bloch equations for the magnetization<sup>14</sup>

$$\frac{\partial \vec{\mathbf{M}}}{\partial t} = \gamma \vec{\mathbf{M}} \times \vec{\mathbf{H}} - \frac{\vec{\mathbf{M}}}{T_2^*} + D \nabla^2 \vec{\mathbf{M}} .$$
 (2)

As suggested by Kaplan,<sup>15</sup> the modified Bloch equation, combined with the Maxwell equations, determines the behavior of the fields within the metal. If Ohm's law is assumed and the displacement current neglected, and if the external field H is normal to the specimen surface, these coupled equations describe two types of propagating waves, characterized by the wave vectors

$$k_1^2 = (2i/\delta^2)(1 + 4\pi\chi\gamma HT_2^*\delta_2/2DT_2^*),$$
  

$$k_2^2 = [1 + i\gamma(H_0 - H)T_2^*]/DT_2^*.$$
(3)

The expansion has been carried out to lowest order in  $\delta^2/2DT_2^*$ , where  $\delta$  is the classical skin depth  $\delta \equiv (c^2/2\pi\sigma\omega_0)^{1/2}$ . The experiment is assumed to be carried out at a fixed frequency  $\omega_0$ , which defines a resonant field  $H_0 \equiv \omega_0/\gamma$ . The first wave vector describes the normal skin effect, slightly modified by the magnetization. The second propagation mode, resonant at  $H = H_0$ , describes conduction-electron spin-resonance transmission and is characterized by an attenuation length  $\delta_s^*$  $\equiv (2DT_2^*)^{1/2}$  which, under certain conditions, may be many times the classical skin depth. The microwave power transmitted through a specimen is found by considering both modes of propagation, subject to appropriate boundary conditions at the two surfaces. In addition to the continuity requirements on tangential  $\vec{E}$  and tangential  $\vec{H}$ , it is necessary to assume a boundary condition on  $\vec{M}$ . The simplest assumption is that there is no surface relaxation, in which case the boundary condition<sup>16</sup> is

$$\hat{n} \cdot \nabla \vec{\mathbf{M}} = 0 \tag{4}$$

where  $\hat{n}$  is the inward unit normal at the surface of the specimen. There is strong evidence that surface relaxation is important for our measurements on sodium. A detailed treatment of surface relaxation effects on the transmission signal is presented in an accompanying paper,<sup>17</sup> whose results will be used here in the analysis of our data.

If there is no surface relaxation, the complex amplitude of the wave transmitted near resonance through a specimen of thickness d is found to be proportional to

$$S = 1/k_2 \sinh k_2 d . \tag{5}$$

Experimentally, this transmitted field is mixed with a coherent microwave field whose phase  $\phi$  may be adjusted. The resulting signal is

$$V = \operatorname{Re}(S) \cos\phi + \operatorname{Im}(S) \sin\phi . \tag{6}$$

Although different choices of phase may be useful, it is most convenient to work with symmetric signals, obtained by setting  $\phi = 0$ , in which case

$$V_{\rm s} = {\rm Re}({\rm S}) \ . \tag{7}$$

For the thick samples characteristic of many of our measurements,  $d \gg \delta_s^*$ , we find

$$V = \operatorname{Re}(-2e^{-k_{2}d}/k_{2})$$
  
= -2(k\_{r}\cos k\_{i}d - k\_{i}\sin k\_{i}d)e^{-k\_{r}d}/(k\_{r}^{2} + k\_{i}^{2}), \qquad (8)

where  $k_i$  and  $k_i$  are the real and imaginary parts, respectively, of  $k_2$ , given by

$$k_{r} = \left\{ \left[ 1 + \gamma^{2} T_{2}^{*2} (H_{0} - H)^{2} \right]^{1/2} + 1 \right\}^{1/2} / \delta_{s}^{*},$$

$$k_{i} = \operatorname{sgn}(H_{0} - H) \left\{ \left[ 1 + \gamma^{2} T_{2}^{*2} (H_{0} - H)^{2} \right]^{1/2} - 1 \right\}^{1/2} / \delta_{s}^{*}.$$
(9)

At resonance,  $k_2$  is real, so the on-resonance signal is

$$V_0 = 1/k_0 \sinh k_0 d , \qquad (10)$$

where  $k_0 \equiv \sqrt{2} / \delta_s^x$ . Thus, subject to the specific assumptions about surface relaxation, a measurement of the on-resonance signal intensity as a function of specimen thickness should permit a straightforward determination of  $\delta_s^x$ . To determine the diffusion constant D, it is also necessary to

know  $T_2^*$ , which requires that the shape of the entire resonance line be observed.

# **B.** Interacting electrons

For interacting electrons, the transport of magnetization cannot, in general, be described so simply. Platzman and Wolf<sup>13</sup> have applied the results of Fermi-liquid theory, appropriately modified for charged systems, to the calculation of spin transport coefficients. They have shown that, under conditions satisfied by our experiments, the only change required is to replace D in the diffusion-modified Bloch equations with  $D^*$ , given by

$$D^* = \frac{1}{3} V_F^2 \tau (1 + B_0) (1 + B_1) / (1 + i X \omega_0 \tau) , \qquad (11)$$

where  $X \equiv (B_1 - B_0)/(1 + B_0)$ . Spin-wave measurements<sup>7</sup> have demonstrated that  $B_1 \ll B_0$  in sodium, potassium, and rubidium. Although there are no comparable measurements in lithium, we assume that this is also the case. Our measurements are, in addition, carried out under conditions for which  $\omega_0 \tau \ll 1$ . We may, therefore, write

$$D^* = \frac{1}{3} V_F^2 \tau (1 + B_0) , \qquad (12)$$

where both  $V_F$  and  $\tau$  must include many-body effects.

Expressing  $D^*$  in terms of the measured quantities  $\delta_s^*$  and  $T_2^*$  we may write

$$1 + B_0 = \frac{3}{2} \delta_s^{*2} V_F^2 \tau / T_2^* , \qquad (13)$$

which, substituted into Eq. (1), gives for the susceptibility enhancement,

$$\frac{\chi}{\chi^0} = \frac{2}{3} \frac{T_2^* V_F^2 \tau}{\delta_s^{*2}} \frac{m^*}{m} \,. \tag{14}$$

Written in this form, evaluation of  $\chi/\chi^0$  from our measurements appears to require a knowledge of  $m^*/m$ , as well as values of  $V_F$  and  $\tau$  that include many-body effects. Fredkin has shown, however, using the results of Fermi-liquid theory for a spherically symmetric magnetic system,<sup>12</sup> that one may write  $V_F^2 \tau m^*/m = (V_F^0)^2 \tau^0$ , where  $V_F^0$  and  $\tau^0$  are the values calculated for free electrons. Using Fredkin's result and the free-electron expressions for  $V_F^0$  and  $\tau^0$ , one then has

$$\frac{\chi}{\chi^0} = \frac{2}{3} \frac{T_2^*}{\delta_s^{*2}} V_F^{02} \tau^0$$
  
=  $\frac{2}{3} (3\pi^2)^{2/3} \frac{h^2}{m_0 e^2} \frac{T_2^*}{\delta_s^{*2} \rho n^{1/3}} .$  (15)

By treating the data in this way one need not calculate a Fermi-liquid parameter explicitly, nor is it necessary to know  $m^*/m$ . In addition to our measurements of  $T_2^*$  and  $\delta_s^*$ , measured values of the dc resistivity  $\rho$  and of the electron density nare required. 890

# C. Surface relaxation effects

Line shapes derived assuming no surface relaxation appear to describe our room-temperature measurements in lithium, but do not describe satisfactorily our results in sodium at 77 K. We summarize here the pertinent results from an accompanying paper<sup>17</sup> in which the effects of surface relaxation are discussed in detail. The boundary condition, Eq. (4), on the magnetization is replaced by

$$\hat{n} \cdot \nabla \vec{\mathbf{M}} = g \vec{\mathbf{M}}, \tag{16}$$

where  $g \equiv 3\epsilon/4\Lambda$  and  $\Lambda$  and  $\epsilon$  are, respectively, the mean free path and the probability of a spin disorientation upon collision with the surface. For conditions satisfied by our experiments, the complex amplitude of the transmitted wave can be written as

$$S = \frac{k_2}{(k_2^2 + g^2)\sinh k_2 d + 2gk_2\cosh k_2 d} .$$
(17)

The real part of S is symmetric about  $H_0$ , so the choice  $\phi = 0$  in Eq. (6) again leads to a detected signal that is symmetric. For thick samples, the symmetric detected signal is

$$V = \frac{2(k_r^2 + k_i^2)[k_i \sin k_i d - (k_r + 2g) \cos k_i d]e^{-k_r d}}{[(k_r + g)^2 + k_i^2]^2} , \quad (18)$$

an expression that reduces to Eq. (8) when g=0. If the surface relaxation is small  $(g/k_2 \ll 1)$ , this may be written, to first order in  $g/k_2$ , as

$$V = \frac{2e^{-k_r d} [k_i \sin k_i d - (k_r + 2g) \cos k_i d]}{k_r^2 + k_i^2 + 4gk_r} .$$
(19)

At resonance S is real, so that  $V_0 = \operatorname{Re}(S_0) = S_0$ , and the general result for the on-resonance symmetric signal may be found from Eq. (17) to be

$$V_{0} = \frac{k_{0}}{(k_{0}^{2} + g^{2}) \sinh k_{0}d + 2gk_{0} \cosh k_{0}d},$$
 (20)

with  $k_0$  defined as before. In the presence of surface relaxation  $k_0$  cannot be found from the onresonance measurements unless one has a value of g, obtained from measurements of the shape of the entire line. However, if  $g/k_0 \ll 1$ , Eq. (20) simplifies to

$$V_0 \approx \frac{k_0}{k_0^2 \sinh k_0 d + 2gk_0 \cosh k_0 d} , \qquad (21)$$

for samples of arbitrary thickness. For thick specimens and arbitrary  $g/k_o$ , one finds instead that

$$V_0 \approx k_0 e^{-k_0 d} / (k_0 + g)^2$$
 (22)

Thus, in the thick specimen regime the surface relaxation contribution is not affected by the

specimen thickness, but enters only through a multiplicative factor that is the same for all specimens. It is assumed, of course, that g will not vary from specimen to specimen, and this is a potential source of trouble with this method.

## **III. EXPERIMENTAL**

## A. Specimen preparation

Bulk sodium was generously supplied to us by C. E. Taylor of Lawrence Radiation Laboratory. A rough measurement in our laboratory gave a bulk resistivity ratio  $\rho(300)/\rho(4.2)$  of better than 4000. Absorption lines as narrow as 80 mG are observed in this material, and we have observed the transmission spin waves first seen by Dunifer et al.<sup>7</sup> Reactor grade lithium of nominal 99.983% purity was bought from the Foote Mineral Company. Thin samples were made by rolling small lumps of the metal in a stainless-steel rolling pack consisting of two stacked rectangular stainless-steel plates, welded together along one edge. Two sheets of carefully cleaned polyethylene were placed between the stainless-steel plates; the lump of metal, covered with mineral oil, was placed between the polyethylene sheets, and the entire pack was passed several times through a hand rolling mill, gradually reducing the metal to the desired thickness. Specimens approximately 1 cm square were cut from the rolled sheet. The thickness of the specimen was determined by measuring with a precision micrometer the thickness at several places around the edge of the hole left after the specimen was removed. The average of these measurements gave the average specimen thickness to  $\pm 2.5 \ \mu m$ . A systematic variation in thickness of  $\approx 4 \ \mu m$  across the 1-cm specimen width could be correlated with the slight V shape of the rolling pack. Since the holes in the transmitter and receiver cavities are 5 mm in diameter, the variation in thickness across the region through which transmission is observed is only about 2  $\mu$ m. Immediately after being cut, the specimens were mounted between the transmitter and receiver cavities, where they were protected on both sides by windows made either of C-8 epoxy or of G. E. "Lucalox".

## B. Transmission spectrometer

The X-band transmission spectrometer was essentially the same as that described previously by one of us,<sup>18</sup> and similar to others described in the literature.<sup>19</sup> A block diagram is shown in Fig. 1. Since accurate intensity measurements were essential, the sensitivity of the spectrometer was carefully calibrated before each transmission



FIG. 1. X-band transmission spectrometer with fully coherent superheterodyne detection. CRT: cathode-ray tube. AFC: automatic frequency control.

run by leaking a small amount of calibrating power into the receiver through an open microwave switch and a precision attenuator. The overall attenuation of this combination was accurately reproducible, and was set at about 130 dB to give a calibrating signal comparable in magnitude to the transmission signal at resonance. The lock-in amplifier output from this calibrating signal was used to normalize the measured intensity for each specimen.

# C. Data analysis

Although the on-resonance intensity as a function of specimen thickness is sufficient to determine  $\delta_s^*$ , additional details of the line shape are needed to determine  $T_2^*$  and g. For thick specimens, the symmetric detected signal, given by either Eq. (8) or Eq. (18), has, in addition to the central peak, prominent side lobes corresponding to a transmitted wave shifted by  $\pi$  with respect to the central peak. The central-peak to side-lobe ratio A/B depends only on the ratio  $\delta_s^*/d$ , and is a useful datum, together with  $\Delta H$ , the full width at half-maximum of the central peak, in determining  $T_2^*$  and g. These quantities are indicated in Fig. 2, which shows a representative thick-specimen symmetric line, assuming no surface relaxation.

Measured values of central-peak intensity as a function of thickness were first fitted by least squares to Eq. (10), assuming no surface relaxation, in order to determine a first approximation to  $\delta_s^*$ . The value of  $\delta_s^*$  thus determined was used as input to a computer program that fit the observed line widths as a function of specimen thickness to the real part of Eq. (17) by varying  $T_2^*$  and g. The resulting value of g was then used to fit the on-resonance intensities in Eq. (20), which includes surface relaxation effects, to determine a new value of  $\delta_s^*$ . Although self-consistent values of  $\delta_s^*$ , g, and  $T_2^*$  could have been determined by successive iterations of this procedure, it was not necessary to do so because we



FIG. 2. Symmetric resonance transmission line shape, Re (S), where S is given by Eq. (5). The detected signal is plotted as a function of the dimensionless variable  $\gamma T_2^*(H-H_0)$  for a thickness  $d=5\delta_s^*$ .

were working in the thick-specimen regime, for which, as shown in Eq. (22), surface relaxation does not affect the thickness dependence of the onresonance signal intensity. In sodium the fit of the linewidth data was sensitive to the choice of g, but in lithium surface relaxation did not appear to be important.

# **IV. RESULTS**

#### A. Lithium

Measurements were made at room temperature on eleven specimens of lithium, ranging in thickness from 60 to 150  $\mu$ m. Since an earlier measurement<sup>18</sup> had estimated  $\delta_s^*$  to be 23  $\mu$ m, all of the measured specimens were expected to be well within the thick-specimen regime, an expectation confirmed by analysis of the on-resonance data, which gave the same value for  $\delta_s^*$ , whether fitted with Eq. (10) or with its thick-specimen limit,  $\exp(-k_0 d)/k_0$ .

Figure 3 is a semilogarithmic plot of on-resonance intensity versus specimen thickness, with a straight-line fit whose slope gives a value of  $\delta_s^* = 23 \pm 2 \ \mu\text{m}$ . From linewidth measurements, we find  $T_2^* = 1.7 \times 10^{-7}$  sec, and negligible surface relaxation. To calculate  $\chi/\chi^0$  from Eq. (15) we have



FIG. 3. On-resonance intensity vs specimen thickness for Li at room temperature. The straight-line fit to the data assumes the thick-specimen dependence given by Eq. (22), and gives a value of  $\delta_s^* = 23 \pm 2 \,\mu$ m.

used a value of  $n = 4.70 \times 10^{28} m^{-3}$  for the electron density, <sup>20</sup> and a resistivity<sup>21</sup> of  $9.32 \times 10^{-8}$  ohm m. The resulting value for the susceptibility enhancement is  $\chi/\chi^0 = 2.86 \pm 0.7$ .

#### **B.** Sodium

Measurements were made at 77 K on eleven specimens of sodium ranging in thickness from 75 to 157  $\mu$ m. Because the thinner specimens were not expected to be well within the thickspecimen regime, all fits of the data to theory used expressions calculated for arbitrary thickness. For example, the first approximation to  $\delta_s^*$  was determined by fitting on-resonance intensities to Eq. (10). This value was then used in fitting the observed linewidths to Eq. (17) to determine  $T_2^*$  and g. The resulting value of g was used in turn in a fit of Eq. (20) to the on-resonance intensities to determine a new value for  $\delta_s^*$ . Since the value of  $\delta_s^*$  was only slightly modified by the introduction of surface relaxation, no further iterations were required. Figure 4 shows the linewidth measurements as a function of specimen thickness. The on-resonance intensities as a function of thickness are shown in Fig. 5, where the fitted curve and the value of  $\delta_{*}^{*}$  are obtained using the value of g derived from the linewidth data of Fig 4. The best value of  $\delta_{\bullet}^*$  is  $44 \pm 5 \ \mu m$ . For the transverse relaxation time we



FIG. 4. Linewidth vs specimen thickness for Na at 77 K. The solid theoretical curve is calculated from the expression for arbitrary thickness using values of  $T_s^*=3.73\times10^{-8}$  sec and  $g/k_0=0.5$ .



FIG. 5. On-resonance transmission intensity in Na at 77 K as a function of specimen thickness. The solid theoretical curve is plotted from Eq. (20) for values of  $\delta_s^*=44 \ \mu m$  and  $g/k_0=0.5$ .



FIG. 6. Linewidth data from Fig. 4 replotted showing error bars. Solid curves are calculated for two limiting sets of  $P (\equiv g/k_0)$  and  $T_s^*$  (measured in units of  $10^{-8}$  sec).

find  $T_2^* = (3.7 \pm 0.5) \times 10^{-8}$  sec, and for the relaxation parameter  $g/k_0 = 0.5 \pm 0.4$ . In Fig. 6 are shown the same linewidth data as shown in Fig. 4, but with theoretical curves corresponding to two extreme combinations of  $T_2^*$  and g. If the onresonance intensity data are fitted with either of these two extreme values of g, the inferred  $\delta_s^*$ does not change appreciably from  $44 \pm 5 \mu m$ . The surface relaxation parameter  $g/k_0 = 0.5$  yields a value for  $\epsilon$ , the probability of a spin flip on collision with the surface, of  $4 \times 10^{-3}$ . Analysis of our spin-resonance transmission data on sodium at 20.4 K, although of little value for a determination of  $\chi/\chi^0$ , yields an estimate of  $\epsilon \approx 10^{-4}$ , in reasonable agreement with the corrected result obtained at this temperature by Wang and Schumacher.<sup>22</sup> These authors have speculated that a true temperature-dependent surface relaxation may be due to surface phonon modes, but we have insufficient data to test their conjecture.

One serious problem with our results in sodium is illustrated in Fig. 7, where the measured lobe ratio A/B is plotted as a function of specimen thickness. Theoretical curves are shown for  $\delta_s^*$ = 44  $\mu$ m and for two values of the surface relaxation parameter  $g/k_0$ . The disagreement with the



FIG. 7. Central peak to side lobe ratio A/B in Na at 77 K as a function of specimen thickness. The theoretical curves are plotted for two values of surface relaxation parameter  $P (\equiv g/k_0)$ .

measured values is serious, particularly for thicknesses  $d \leq 2\delta_s^*$ . Although a determination of  $\delta_s^*$  and  $T_2^*$  does not require fitting of the side-lobe ratio data, our inability to explain the thickness dependence of this ratio does raise doubts about the validity of our analysis.

To calculate the susceptibility enhancement in sodium, we use values for the electron density<sup>20</sup> of  $2.57 \times 10^{28}$  m<sup>-3</sup> and for the resistivity<sup>23</sup> of  $0.805 \times 10^{-8}$  ohm m. The susceptibility enhancement calculated from Eq. (15) is  $\chi/\chi^0 = 2.45 \pm 0.7$ , a result that, because of its large error, is not incompatible with the most recent direct measurement<sup>6</sup> of  $1.67 \pm 0.02$ , but is not deserving of serious comparison.

# V. DISCUSSION

Because our value of  $\chi/\chi^0$  for sodium, apart from its large error limits, differs considerably from other reported values, we shall not undertake a detailed comparison. It may be that a better theory of surface relaxation is required. A more likely explanation is that surface relaxation is adequately treated but that differences in the preparation and handling of the specimens result in a different value of g for each specimen. If that is the case, much of our analysis is inapplicable. Measurements, such as those of Flesner and Schultz, that use only a single specimen are not susceptible to variations in g. Our value of  $T_2^*$  is in reasonable agreement with other measurements, <sup>24</sup> and is not a major source of error.

For lithium, our value of  $\chi/\chi^0$  agrees with other recent measurements<sup>6,8,25</sup> and with the best recent theoretical calculation.<sup>26</sup> Unfortunately, our error bars are so large that they span the entire range of the other reported measurements and include both a recent direct measurement and the measurements of Flesner and Schultz, carried out by methods similar to ours.

Flesner and Schultz have measured  $\chi/\chi^0$  by two methods. One method measures  $\delta_s^*$  and  $T_2^*$  by fitting line shapes for a single specimen as a function of temperature in the regime of completely damped spin waves, and determines  $\chi/\chi^0$ essentially as we do, by using an expression equivalent to Eq. (15). Because they use only a single specimen, their method has a smaller error than ours, and yields a value of  $\chi/\chi^0 = 3.1 \pm 0.3$ . Their other method determines the coefficient  $B_0$ from an analysis of line shape asymmetries in the regime of partially damped spin waves when  $D^*$  is



FIG. 8. Recent experimental and theoretical results for the susceptibility enhancement  $\chi/\chi^0$  in lithium: *P*—present work; WVS—Whiting, VanderVen, and Schumacher (Ref. 6); KSV—Kettler, Shanholtzer, and Vehse (Ref. 25); FS1—Flesner and Schultz, partially resolved spin waves (Ref. 8); FS2—Flesner and Schultz, completely damped spin waves (Ref. 8); VP—theoretical calculation of Vosko, Perdew, and McDonald (Ref. 26).

complex. The value of  $B_0$  inferred from these measurements is dependent upon the choice of  $m^*$ , for which there are two different values derived from specific-heat data.<sup>27,28</sup> The quoted values for the susceptibility enhancement are  $2.93 \pm 0.1$ using an effective-mass ratio of 2.34, and 2.84  $\pm 0.1$  using an effective-mass ratio of 2.21. We believe, since there is no reason to prefer either effective-mass value, that their measured susceptibility enhancement should be quoted as the single value  $2.89 \pm 0.2$ , which reflects more realistically the uncertainty in the effective-mass data.

The results of several recent measurements of  $\chi/\chi^0$  in lithium are summarized in Fig. 8, together with the best recent theoretical value. The results obtained from spin-resonance transmission measurements tend toward somewhat higher values than do the results of direct measurements. We believe that the latter measurements are more reliable, not only because of their smaller error due to scatter in the data, but because their interpretation is not based so heavily on the results of a phenomenological theory.

## ACKNOWLEDGMENTS

We should like to thank Professor R. T. Schumacher for several helpful discussions. This work was supported by a grant from the NSF, and is based on a thesis submitted by C. E. Witt to the Physics Dept. of Carnegie-Mellon University in partial fulfillment of the Ph.D. requirements.

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