Line shapes and A/B and g values of conduction-electron spin resonance in aluminum

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The experimental conduction-electron spin-resonance absorption line shapes in aluminum have been extensively compared to the theory, for temperatures ranging from 4.4 to 327 K. The agreement in the completely anomalous skin-effect region is excellent. It is therefore possible to determine an accurate g value for aluminum. In the normal skin-effect region the discrepancies disappear if the electron mean-free path is taken to be shorter than the one deduced from resistivity measurements.

INTRODUCTION

Conduction-electron spin resonance (CESR) in pure Al has been reported by Schultz *et al.*[1] and Lubzens *et al.*[2] They used a transmission technique (TESR) and determined linewidths and g factors in the temperature range from 1.4 to 107 K. TESR has the advantage of being very specific for conduction electrons. It is however limited in applicability: the sample thickness d should be larger than the skin depth δ and not much larger than the "spin depth" δ_e which is equal to $(2DT_2)^{1/2}$, where D is the diffusion constant and T_2 is the electron-spin relaxation time. This limitation reflects in the range of temperatures and sample thicknesses available for the measurement. The more conventional reflection technique is therefore better adapted to study CESR in all the different cases that are considered in the Dyson[3] theory.

EXPERIMENTAL DETAILS

We used a reflection spectrometer at a frequency of 20.98 GHz, equipped with a cylindrical cavity driven in the TE_{011} mode. Details of the spectrometer, which is specially suited for the detection of broad resonance lines, have been published elsewhere.[4] The samples were in the form of thin platelets with thicknesses of 0.8, 2.4, and 26 μm. They were prepared from very pure[5] (99.9999%) Al by cold rolling followed by electrolytic etching. In a series of measurements of linewidth and g value versus sample thickness, it was checked that samples prepared from a single crystal by spark cutting, grinding, and electropolishing give essentially the same results. The large signal-to-noise ratios needed for reliable fitting of theoretical line shapes were obtained by making stacks of about ten sample plates separated by Teflon sheets. An example of how well the Dyson theory can describe the experimental line is shown in Fig. 1.

EXPERIMENTAL RESULTS AND DISCUSSION

The analysis was done using the expressions obtained from the Dyson theory by Pifer and Magno.[6] The measurements fall into different cases as shown in Table I. The theoretical resonance lines were compared in each case to the corresponding experimental lines, in a magnetic field region extending over at least eight linewidths. This reveals the relevant differences, which are most pronounced in the wings of the lines. It makes possible an accurate evaluation of the asymmetry parameter A/B, as defined in Fig. 1, and tells us the exact magnetic field value H_0, needed for the determination of the g factor; the A/B value is a convenient parameter to compare directly theory with experiment. The results are shown in Figs. 2(a), 2(b), and 2(c).

The shown theoretical values have been calculated

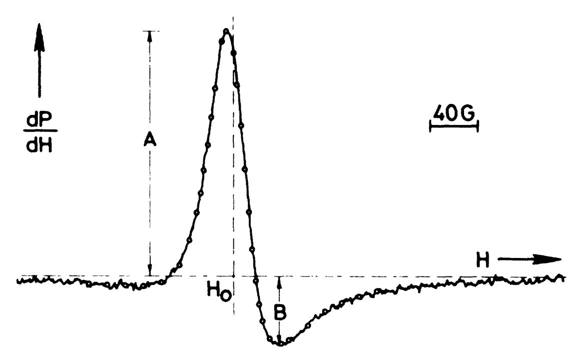

FIG. 1. Typical CESR line shape (derivative of the power absorption) of an aluminum sample with thickness $d = 2.4$ μm at a temperature of 20.5 K. The modulation-field amplitude was less than 0.1 times the linewidth. The dots are points of a theoretical anomalous skin-effect line, with $d/\delta \gg 1$ and $\lambda \gg \delta$ (the A/B value of the line is 3.6). This shows how close the Dyson theory, modified for the anomalous skin effect, fits the experiment.

TABLE I. Subdivision of the experimental conduction-electron spin-resonance region in Al following the Dyson theory, using the sample thickness d and the skin depth δ, and spin depth δ_e deduced from the resistivity measurements.

Condition [a]	Case [a]	d (μm)	Applies to experimental T (K) region [b]
(A) Normal skin effect			
(1) $d/\delta \leq 2$	Very thin	0.8	180–275
(2) $2 \leq d/\delta \leq 2\pi$	Thin	2.4	150–327
(3) $d/\delta \geq 2\pi$			
(I) $d/\delta_e \leq 2\pi$	Intermediate	26	150–160
(II) $d/\delta_e \geq 2\pi$	Thick	26	160–256
(B) Anomalous skin effect			
(1) $d/\delta >> 1$	Thick	0.8	5–25
		2.4	4.4–120
		26	10–150
(2) $d/\delta \lesssim 1$	(No theory	0.8	25–150
	available)	2.4	120–150

[a] See Refs. 3 and 6.
[b] Approximated values.

using the resistivity curve to obtain the electron mean free path λ and using the measured linewidth to obtain the spin-relaxation time T_2.

Our results can be summarized as follows:

(i) In the anomalous skin-effect region at temperatures $T < 60$ K, the theory for $d \gg \delta$ and for diffuse scattering[6] ($p = 0$) of the electrons at the metal surface, describes the experiment very closely. This is shown by the excellent line fitting in Fig. 1 and by the A/B values in Figs. 2(b) and 2(c). Note that the experimental points lie always slightly below the theoretical curve, a fact that has been found in Li also.[6] The data for the 0.8-μm film, shown in Fig. 2(a), cannot be compared to any valid theoretical curve, except perhaps at the very low temperatures (see Table I), where $d/\delta \gg 1$ and here the agreement is quite good.

The large signal-to-noise ratio and the almost-perfect line fitting make it possible to determine the g factor with high accuracy in this low-temperature region. The results are given in Figs. 3(a) and 3(b). There is a rise in g value at the lowest temperatures, which has been reported before[2,7] and was explained as due to exchange interactions between conduction electrons. Note that this rise in g factor at the lowest temperatures depends on the sample thickness as can be seen by comparing Figs. 3(a) and 3(b). This fact is not surprising because the resistivity relaxation time τ gets more influenced by size effects as the sample thickness decreases. It is this relaxation time which is important in explaining the g increase.[2,7] The change in g value disappears above 25 K so that from there on the intrinsic motionally narrowed g factor is obtained and this g value stays constant up to room temperature. Its value is determined as 1.9953 ± 0.0003; this ameliorates the result from TESR measurements which gives a g value of 1.997 ± 0.001.

It has been reported before[7] that there should be an experimentally meaningful g shift above 60 K. As shown in Figs. 3(a) and 3 (b), there is no evidence of this fact anymore. The reasons for the earlier mistakes are the following: our previous signals in thick aluminum samples above 60 K had a poor signal-to-noise ratio, which made it difficult to determine the A/B value. Thus, in order to deduce the g value from the line we used the theoretical A/B value as predicted by the Dyson theory.[3,6] As we show in this work, there are large discrepancies between experimental and theoretical A/B values in the discussed temperature region. This fact misled us in the sense that we applied too small corrections to the g values determined on the top of the resonance line.[6,8] The top g value is higher or equal to the real g value (except in the region where $\pi < d/\delta < 2\pi$, for certain δ/δ_e values), and the correction depends on the A/B value and the linewidth.

(ii) At intermediate temperatures from about 60 K to about 150 K, the skin effect evolves from completely anomalous to normal. The A/B values taken from the experimental lines are much smaller than those obtained from the extrapolation of the anomalous skin-effect theory. The so-called δ/δ_e peak in the A/B curve has almost completely disappeared. It should be noted again that there is

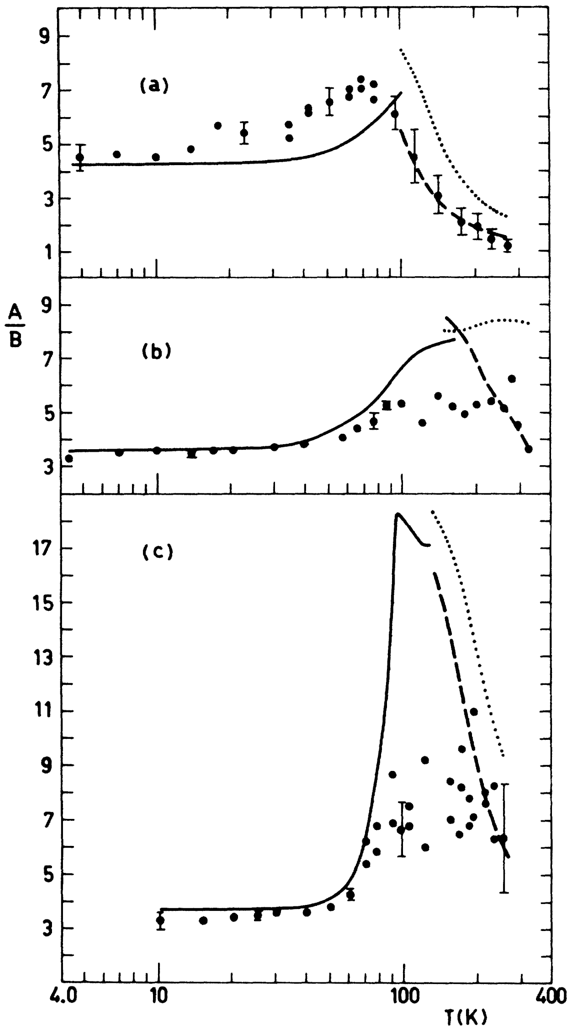

FIG. 2. A/B values vs temperature for the derivative of the CESR power absorption in aluminum samples with thicknesses $d = 0.8\ \mu m$ (a), $2.4\ \mu m$ (b), and $26\ \mu m$ (c). The solid lines give the theoretical A/B values in the anomalous skin-effect region for the thick-film case ($d \gg \delta$) deduced for each sample from the thickness d, the skin depth δ, and the spin depth δ_e. The dotted lines give analogous theoretical A/B values but now for the normal skin-effect region. The dashed lines are also theoretical curves for the normal skin-effect region but the skin depth has been chosen smaller by factors 1.4, 1.9, and 1.3 for the samples with $d = 0.8$, 2.4, and $26\ \mu m$, respectively.

no theoretical A/B curve available for the 0.8-μm film.

(iii) At temperatures of about 150 K to room temperature, being completely in the normal skin-effect region, the A/B values are smaller than those given by the corresponding normal skin-effect

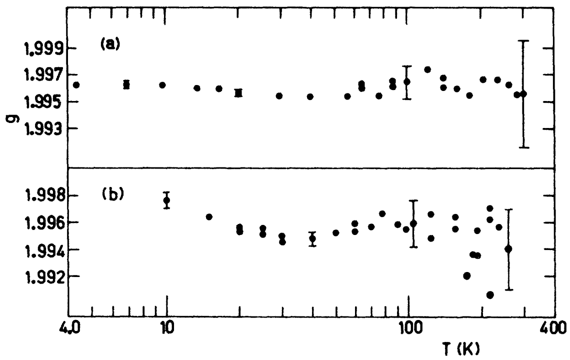

FIG. 3. Temperature dependence of the electronic g factors in aluminum samples with thicknesses $d = 2.4\ \mu m$ (a) and $d = 26\ \mu m$ (b). In the motionally narrowed regime, the g value is determined as 1.9953 ± 0.0003.

theory. There have been previous reports of this effect in the literature for alkali metals and it has been tentatively connected to surface relaxation.[6,8]

Dyson however showed that for the thick case in the normal skin-effect region (the case that surely applies to our 26-μm sample), surface relaxation can change the line shape (and the linewidth) on the condition that a parameter ϵ is large enough, ϵ being the fraction of surface collisions that are spin relaxing. Large enough means $\epsilon \gtrsim (\tau/T_2)^{1/2}$, where τ is the resistivity collision time. It is estimated from measurements of linewidth versus sample thickness,[9] that in Al at liquid-He temperature, ϵ must be of the order of 10^{-4}. The $(\tau/T_2)^{1/2}$ value at temperatures above 150 K is close to 10^{-2}, so that it is difficult to see how the surface relaxation could be involved unless ϵ should be much larger at 150 K than at 4 K. This is not expected from normal models so that we should leave a question mark as to the cause of this discrepancy in A/B values.

The line fittings in the temperature range between 150 K and room temperature can be performed very satisfactorily if one chooses theoretical lines with the right A/B values, and this applies to all the cases shown in Table I and Figs. 2(a), 2(b), and 2(c). These fittings show, however, that there exists a discrepancy between the electron mean free path λ that produces the right A/B value from the theory and the λ that is obtained from bulk resistivity measurements. All the values have to be corrected so that the skin depth $\delta \sim \lambda^{-1/2}$ is to be taken larger in CESR than in resistivity. The corrections to δ are by factors 1.4, 1.9, and 1.3 as shown in Figs. 2(a), 2(b), and 2(c), respectively. A comparable correction was measured in Cu by means of a different experiment by Tisher.[10]

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