Conduction-electron spin resonance in aluminum at 79 GHz*

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Conduction-electron spin resonance in aluminum has been studied by the microwave transmission technique at 79 GHz over the temperature range 4–80 K. At low temperatures the g value and linewidth show the same type of anomalous temperature dependence as reported earlier at 35 GHz. At high temperatures, where the linewidth reflects the spin-flip scattering rate from thermal phonons, a strong and unexpected frequency dependence is observed with the linewidth being proportional to the applied frequency.

I. INTRODUCTION

In this paper we report our measurement of conduction-electron spin resonance (CESR) in high-purity aluminum at a frequency of 79 GHz. Earlier observations at 35 GHz by Lubzens, Shanabarger, and Schultz[1] revealed an anomalous behavior of the temperature dependence of the CESR g value and linewidth. This was interpreted by Fredkin and Freedman[2,3] in terms of a complex model containing g anisotropy around the Fermi surface, motional narrowing of the CESR, and a relatively strong exchange coupling between the conduction electrons. By comparing data at both 9.2 and 35 GHz with the theoretical model Lubzens *et al.* were able to obtain a self-consistent fit and determine approximate values for σ_g the rms spread of g values, and B a many-body exchange parameter. However, very recently Lubzens and Schultz[4] have reported additional measurements on aluminum at 1.27 GHz which do not appear to be in agreement with the earlier results and predictions of the theory. Our 79-GHz measurements provide yet another test of this theory in the crucial frequency region where electron-electron exchange effects are expected to predominate.

According to the model of Fredkin and Freedman, at relatively low temperatures where spin-flip scattering of the conduction electrons by thermal phonons does not contribute appreciably to the CESR linewidth, there can be two distinct temperature regimes in which the nature of the CESR signals will be quite different from each other. In the higher-temperature limit of this theory, where momentum scattering of the electrons from thermal phonons is still sufficiently rapid, exchange coupling of the electrons is relatively unimportant and the CESR signal displays the average g value on the Fermi surface and is *motionally* narrowed by rapid scattering of the electrons. In the lower-temperature limit of the theory, the momentum scattering rate decreases, and motional narrowing of the CESR signal becomes relatively unimportant. In this regime the exchange coupling between the spins leads to a collective mode in which all the conduction electrons precess together at the same Larmor frequency. This collective mode is characterized by both *another* form of linewidth narrowing (exchange narrowing) and a shift in g value from that displayed at higher temperatures. Fredkin and Freedman characterize the two regimes by the parameter X, where

$$X = B(1+B)^{-1}\omega\tau \, , \qquad (1)$$

ω is the applied frequency, and τ is the momentum scattering time. When $X \ll 1$, one is in the higher-temperature regime in which motional narrowing plays the dominant role; when $X \gg 1$, the exchange coupling is dominant and the collective mode is established.

The interpretation of previous experimental investigations has been that either motional narrowing has played the dominant role, or else at the highest frequencies employed (35 GHz) the roles of motional narrowing and exchange coupling were only comparable ($X \approx 1.0$). The distinct advantage of our measurements at the higher frequency of 79 GHz is that on the basis of the lower-frequency results we should be able to achieve $X \gg 1$ and thus provide an additional check on the theory in that regime where exchange coupling will play the major role in determining the CESR properties.

II. EXPERIMENTAL TECHNIQUE

The CESR measurements were made over the temperature range 4–80 K using the microwave transmission technique[5-7] at a frequency of 79 GHz. With this technique one monitors microwave power carried through the sample (from one surface to the other) by the diffusion of resonant electrons. The primary advantage of the transmission technique is that it discriminates very strongly against background resonances due to impurities present. The dc magnetic field was applied perpendicular to the surface of the samples which were in the form of thin plates (both single crystal and polycrystalline) whose thicknesses ranged from 0.027 to 0.30 mm. The aluminum[8] had a typical purity of 99.9999% and in the bulk displayed an electrical resistivity ratio (between room temperature and liquid-helium temperature) as high as 20 000. In the thinner samples the resistivity ratios were limited by surface scattering of the conduction electrons.

Figure 1 shows a block diagram of our microwave transmission spectrometer. A klystron tube is used to generate approximately 500 mW of microwave power, which proceeds through a series of isolators, a variable attenuator, and a modulator before entering the microwave cavity system which is centered in a superconducting solenoid capable of providing a dc magnetic field of up to 60 kG. The input power is square-wave (amplitude) modulated ($\approx$20 kHz) so that phase-sensitive detection can be used in the final stage of signal recovery to optimize signal-to-noise ratio. The sample itself makes up a common wall between two microwave cavities. The top

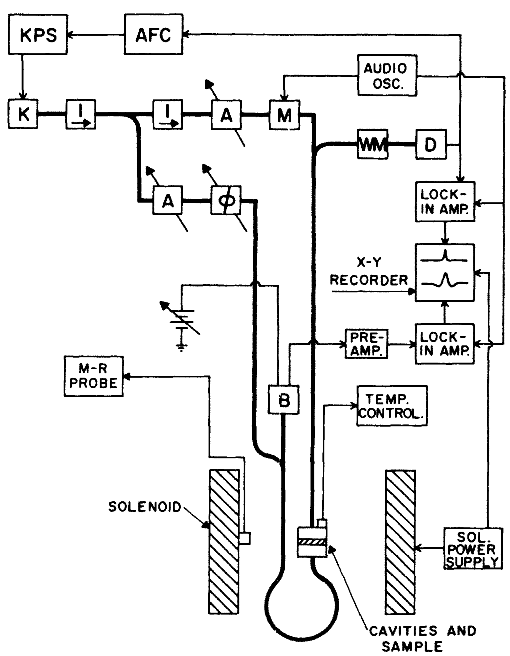

FIG. 1. Block diagram of the 79-GHz microwave transmission spectrometer. Components of the spectrometer include: K—klystron, source of the microwave power; I—microwave isolators; A—variable microwave attenuators; ϕ—variable microwave phase shifter; M—microwave modulator; WM—microwave wavemeter for frequency determinations; D—microwave detector diode; B—InSb bolometer; KPS—klystron power supply; AFC—automatic frequency control circuitry; and M-R probe—magnetoresistance probe to monitor the magnetic field strength.

cavity, called the transmit cavity, couples the microwave power to the sample, while the bottom cavity, called the receive cavity, picks up any power transmitted through the sample. Both cavities are fully tunable so that their resonant frequencies can be matched to that of the klystron.

Any transmitted signal then proceeds around a loop and up to an ultrasensitive detector, a liquid-helium-cooled hot-electron InSb bolometer. Under optimum operating conditions this bolometer can detect a 10^{-21}-W signal with a signal-to-noise ratio of 1 : 1 within a 1-Hz bandwidth. For maximum sensitivity we operate the bolometer as a mixer. This is possible because of its highly nonlinear I-V characteristics.[9] In the mixing mode a small fraction of *unmodulated* power from the original klystron is mixed with the signal before it arrives at the bolometer. This reference or local oscillator power is typically several orders of magnitude larger than the signal itself and as a consequence the output of the bolometer is proportional to only that component of the signal which is in phase with the microwave reference. The phase of the reference, however, is variable and one can consequently adjust this phase to obtain either symmetric (χ''-type) signals or antisymmetric (χ'-type) signals. The output impedance of the bolometer is matched to that of a preamplifier by means of a simple tuned LC circuit (tuned to the 20-kHz modulation frequency). The

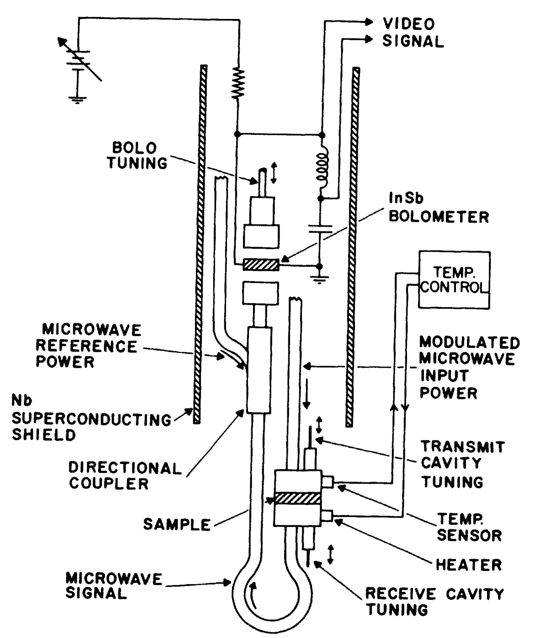

FIG. 2. Close-up view showing that portion of the spectrometer maintained at cryogenic temperatures. Included in this region are the microwave cavities and sample, the InSb bolometer with coupling circuitry, and the superconducting niobium shield.

bolometer and its impedance-matching circuitry are maintained at liquid-helium temperatures and surrounded by a superconducting niobium shield to screen out any dc magnetic field or ac pickup. To make full use of the sensitivity of the bolometer, it is necessary to have a high degree of isolation between the input power to the transmit cavity and the detector system so that the only *modulated* microwave power that appears at the bolometer is that which is carried across the sample by diffusing electrons. Typically, 180 dB of isolation is required.

The part of the spectrometer maintained at cryogenic temperatures is shown in Fig. 2. This portion of the spectrometer is situated in a vacuum can which is surrounded by liquid helium. Thermal contact with the liquid helium is maintained by a small quantity of helium exchange gas in the can. The sample and cavities themselves are thermally isolated from the rest of the system by a thin-walled stainless-steel waveguide so that their temperature can be varied. A temperature control system consisting of heaters and sensors is connected to the cavities permitting their temperature to be varied over the range of 1.2–100 K.

When measuring g values, a g marker consisting of neutron-irradiated LiF ($g = 2.002\,32$)[10] is placed in the transmit cavity and its ESR signal monitored by the usual reflection techniques using a room-temperature Shottky detector diode. By simultaneously recording this signal along with the transmission signal from the aluminum sample, an accurate determination of the CESR g value was made.

III. RESULTS AND DISCUSSION

Figure 3 displays typical CESR traces at several temperatures with the phase of the microwave reference chosen to produce a symmetric signal. The low-temperature base line is complicated by extra transmission signals produced by the cyclotron motion of the electrons. At higher temperatures the CESR develops extra oscillations or lobes, which result from the changing phase of the transmitted signal as one sweeps away from the center of the line.[11]

Figure 4 is a plot of the measured g value as a function of temperature for samples of four different thicknesses. A relatively strong temperature dependence of the g value begins around 30 K and extends down to about 5 K. The g value remains fairly constant above 30 K although another increase, which becomes significantly larger than the experimental uncertainties, is observed at temperatures above 50 K. A similar increase in the g value of aluminum at high temperatures has

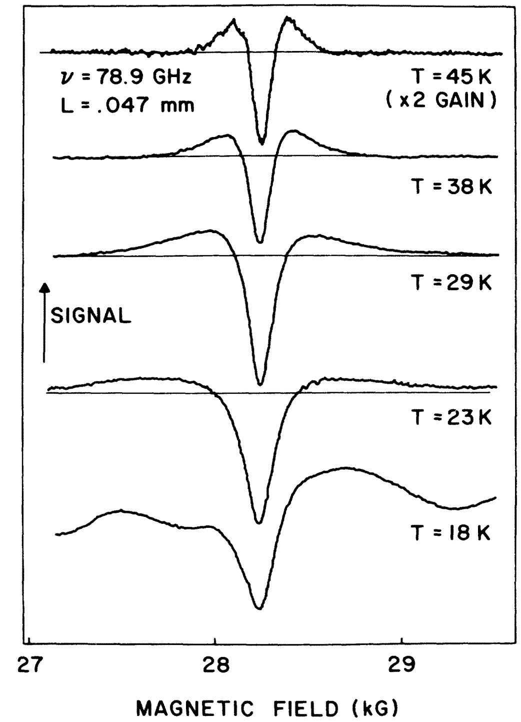

FIG. 3. Typical CESR traces for a high-purity aluminum plate at a variety of temperatures. The electrical resistivity ratio of the sample is ≈2000 and is limited by surface scattering of the electrons.

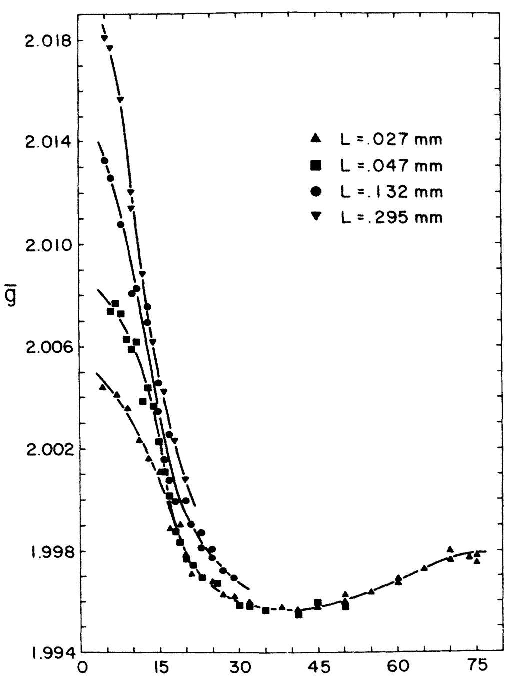

FIG. 4. The temperature dependence of the observed CESR g value ($\bar{g}$) for aluminum plates of four different thicknesses. Typical uncertainties in a g-value measurement are ±0.0005.

recently been reported by Janssens *et al.*,[12] who observed the CESR signal by reflection spectroscopy at 21 GHz. Although the quoted uncertainties in their g-value determinations are several times larger than ours, their data appear to show the same overall increase with temperature, with approximately the same slope, as we observe over the range 50–80 K. Between 30 and 50 K we obtain $g = 1.9960 \pm 0.0005$, which agrees quite well with the g-value measurements[1,4] at 1.27 and 9.2 GHz and also those at 35 GHz for temperatures above 20 K. The behavior at temperatures below 30 K is very similar to that observed at 35 GHz.

According to the model of Fredkin and Freedman the shift in g value at low temperature will depend upon X and should be given by

$$\Delta g = \Delta g_{\max}[X^2/(1+X^2)] \,, \tag{2}$$

where

$$\Delta g_{\max} = (1+B)^2 \sigma_g^2 / Bg \tag{3}$$

and g is the average g value on the Fermi surface. The resistivity ratios of the four samples are limited by surface scattering and in order of increasing thickness are approximately 1000, 2000, 5000, and 10 000. This means at low temperatures the parameter X is expected to vary appreciably among the four samples, being smallest for the thinnest and largest for the thickest one. With this in mind the observed g shifts at low temperature are consistent with Eq. (2) for each of the four samples. For the thickest sample at the lowest temperature Δg is expected to be very close to $\Delta g_{\max}$, which is thus given by $\Delta g_{\max}(79\text{ GHz}) = 0.022$. According to Eq. (3) $\Delta g_{\max}$ should be independent of frequency; however, the value we obtain is somewhat larger than that obtained at 35 GHz: $\Delta g_{\max}(35\text{ GHz}) = 0.015 \pm 0.003$.

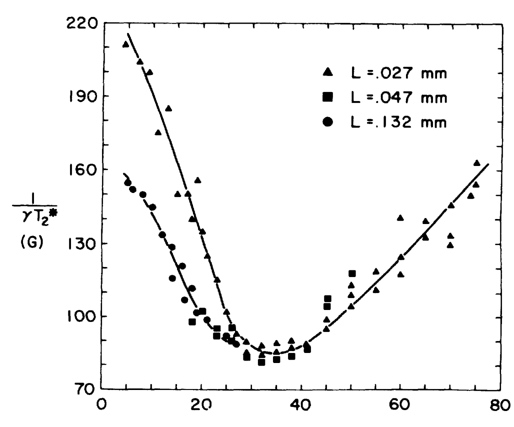

FIG. 5. The temperature dependence of the corrected linewidth (see Ref. 11) for aluminum plates of three different thicknesses. Near the linewidth minimum the uncertainties are typically ±5% rising to ±10% at both the high- and low-temperature extremes.

In Fig. 5 the temperature dependence of the corrected linewidth[11] $1/\gamma T_2^*$ is plotted. In aluminum, T_2^* is expected to contain contributions from both true spin-lattice relaxation and also the g anisotropy around the Fermi surface. The 79-GHz data show the same general behavior of $1/\gamma T_2^*$ as observed at 35 GHz, with the broadening at low temperature being substantially stronger. The linewidth shows a distinct minimum at 35 K and then increases with increasing temperature presumably due to an increase in the spin-flip scattering from thermal phonons. The increase in linewidth at lower temperatures according to the model is due to the reduction in motional narrowing that takes place as the momentum scattering of the electrons is reduced.

In addition to the larger $\Delta g_{\max}$ we observe, even more serious problems are encountered when we attempt a quantitative fit of our linewidth data to the theory. According to the model the contribution to the linewidth from the g anisotropy is given by

$$\Delta H_a = \Delta H_{\max}[2X/(1+X^2)] \,, \tag{4}$$

where

$$\Delta H_{\max} = (1+B)^2 \sigma_g^2 \,\omega / 2g^2 B\gamma \tag{5}$$

and γ is the average value of the electronic gyromagnetic ratio around the Fermi surface. According to Eq. (4) the linewidth at low temperature should have a maximum value when $X = 1.0$. When X grows smaller than 1.0 the line should narrow because of motional narrowing. When X grows larger than 1.0 the line should again narrow because of the exchange coupling between the spins (exchange narrowing). The fit between theory and experiment at 35 GHz for a 0.040-mm-thick sample with a resistance ratio of ≈ 1600 indicated that $X(1.3\text{ K}, 35\text{ GHz}) \approx 1.2$, meaning that the line should have just started to narrow again at the lowest temperatures. As mentioned before, the primary advantage of our higher frequency is that we can obtain $X \gg 1$, a regime in which exchange effects play the dominant role. At 79 GHz and with the thicker samples of Fig. 5 we would expect $X \approx 3-5$ at the lowest temperature, meaning the linewidth should have gone through a peak upon cooling below 15 K or so. No such peak is seen in Fig. 5, nor has such a peak ever been seen for any samples we have run.

Another discrepancy is the size of the linewidth at low temperatures. According to Eqs. (4) and (5) the maximum value the linewidth can go through is $\Delta H_{\max}$, which is linearly dependent upon the applied frequency. Since $\Delta H_{\max}$ at 35 GHz for the aforementioned sample was determined to be 47 G,

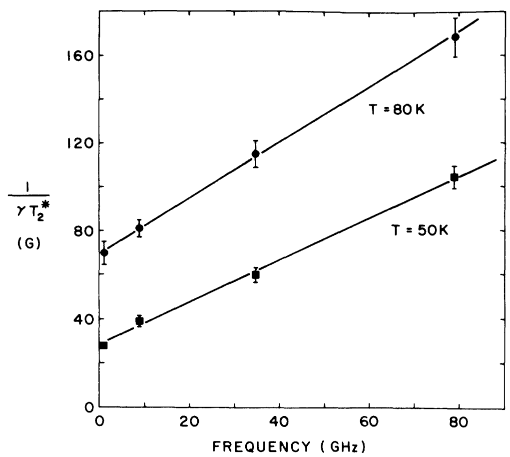

FIG. 6. Frequency dependence of the CESR linewidth at high temperatures where the linewidth is dominated by spin-flip scattering from thermal phonons.

we would not expect to see any linewidths greater than about 106 G at 79 GHz. As can be seen, however, our linewidths at low temperature are appreciably greater than this, with one sample having a linewidth of over 200 G. The linewidths are also somewhat larger than the 130 G that one would extrapolate from the linear frequency dependence observed in Ref. 4.

Still another difficulty is that the largest linewidths are found for the thinnest samples. Until one goes beyond $X=1$ (clearly not seen for any sample), the broadest lines should correspond to those samples with the largest X values. Those samples with the largest resistivity ratios (thickest samples) would be expected to have the largest X values at low temperature, and this is in agreement with the observed g shifts (see Fig. 4). However, as can be seen in Fig. 5 it is the thinnest samples, not the thickest, which have the broadest lines at low temperature.

Another important feature of the linewidth data is its temperature dependence at relatively high temperature ($T \gtrsim 50$ K), where the linewidth is expected to be dominated by spin-flip scattering of the conduction electrons due to their interaction with thermal phonons. The coupling of the electron spins with the lattice is thought to be due to a weak spin-orbit interaction, which is *not* expected to be frequency dependent.[13,14] That is, at higher temperatures the linewidths measured at different frequencies are expected to converge to a single temperature-dependent value as the temperature is raised. Studies of copper and silver[1,4] have shown this to be the case.

However, the CESR linewidth for aluminum shows a distinct frequency dependence in the high-temperature regime. As noted by Lubzens and co-workers[1,4] the linewidths measured at 1.27, 9.2, and 35 GHz do not converge in this limit, but rather the higher-frequency linewidths remain broader. This behavior continues at 79 GHz and is substantially stronger. Figure 6 shows a plot of $1/\gamma T_2^*$ versus frequency for fixed temperatures of 50 and 80 K for our data and that from Refs. 1 and 4. At both temperatures we obtain an excellent straight-line fit strongly suggesting a linear frequency dependence to the phonon-induced spin-flip scattering rate.

In summary, the CESR linewidth of aluminum displays several anomalous features. The low-temperature behavior does not appear to be properly described by the model of Fredkin and Freedman. Possibly, higher-order terms in their expansion may improve the fit between experiment and theory. The frequency dependence of the linewidth at higher temperatures clearly points out the need for a theoretical reexamination of the nature of the phonon contribution to spin relaxation in aluminum. Our measured g values show better agreement with the 35-GHz data and the predictions of the theory, although our value for Δg_{max} is somewhat larger. The g shift for temperatures higher than 50 K remains unexplained at this time.

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