Observation of an Anomalous Frequency Dependence of the Conduction-Electron Spin Resonance in Al†

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New data at 1.27 GHz for conduction-electron spin resonance (CESR) in aluminum are reported. When analyzed in conjunction with our previous data at 9.2 and 35 GHz, we are unable to interpret all the CESR data in terms of existing theory which incorporates g anisotropy and many-body effects, and assumes as frequency independent the phonon, impurity, and surface contributions to the spin relaxation. Analogous data for copper are consistent with the theory.

In a previous Letter[1] (LSS) we reported our observation of a frequency dependence of the transmission-electron-spin-resonance (TESR) signals for Al, Cu, and Ag. We suggested that this frequency dependence was a consequence of g anisotropy over the Fermi surface, and interpreted the data in terms of a theory developed by Fredkin and Freedman[2] (FF), which incorporated both motional narrowing and many-body effects.[3] The analysis of the data at 35 and 9.2 GHz yielded values for the rms spread of the g value and a measure of the strength of the many-body interaction. While the results for Cu and Ag were in reasonable agreement with other measurements, we noted that the rms spread of g for Al was surprisingly large (~4%) and suggested the need for an extension of the measurements to additional frequencies.

We have recently completed the construction of a new spectrometer which operates at a frequency of 1.27 GHz, and have obtained TESR spectra for single-crystal samples of Cu, Ag, and Al at cryogenic temperatures.[4] In Fig. 1 we present the conduction-electron-spin-relaxation (CESR) rate or linewidth ($\Delta H = 1/\gamma T_2{}^*$), as a function of temperature for 35, 9.2, and 1.27 GHz.[5] The data for 35 and 9.2 GHz are similar to those presented in LSS. We note that the 1.27-GHz data are qualitatively similar to those at 9.2 GHz, but the residual linewidth at low temperature (ΔH_R) is only ~5 G for the 0.004-cm sample. (We attribute the slightly narrower line of the 0.018-cm sample to a decrease in surface relaxation.) The g value for Al at 1.27 GHz was 1.996 ± 0.001 over the temperature range of our measurements (1.3–40 K) which is in agreement with the 9.2-GHz value.

In LSS we analyzed the difference in linewidth and g values between 35 and 9.2 GHz utilizing Eqs. (1) and (2) for a 0.004-cm-thick sample. We made the explicit assumptions that the impurity, surface, and phonon contributions to the linewidth were additive and frequency independent. Values of $\Delta H_{\max}$, $\Delta g_{\max}$, and X were chosen which represented a reasonable fit to the data over the temperature range 1.4–20 K. When we utilized these parameters in conjunction with the FF theory to calculate the frequency dependence of the residual linewidth [under the condition $B/(1+B) \gg \sigma_g/g$], we obtained the dotted curve

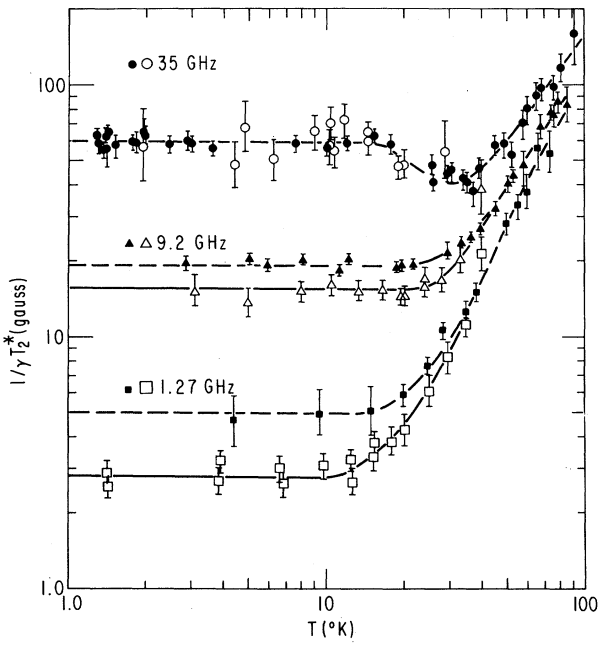

FIG. 1. Temperature dependence of the CESR linewidth, $\Delta H = 1/\gamma T_2{}^*$, for single-crystal Al at 1.27, 9.2, and 35 GHz. The open symbols represent data taken on the same sample, which was 0.018 cm thick, and had a resistivity ratio (room temperature/4.2 K) ~4000. The solid symbols are for analogous data taken on 0.004-cm-thick samples, where the resistivity ratio is ~1600. We attribute the difference in linewidth (~2 G) between thick and thin samples to surface relaxation.

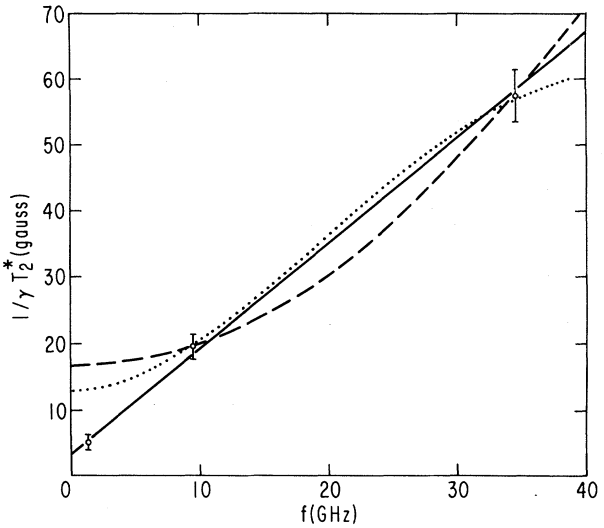

FIG. 2. Frequency dependence of the residual CESR linewidth at 1.4 K, $\Delta H_R = 1/\gamma T_2^*$. The experimental points are for a sample 0.004 cm thick. The theoretical curves were fitted to the data at 9.2 and 35 GHz and then extrapolated to low frequency. The dashed curve assumes ΔH_R may be expressed as the sum of a constant plus a frequency-dependent term based upon complete motional narrowing of the g anisotropy. The dotted curve assumes that the frequency-dependent term is to be expressed by the FF theory (Ref. 2) which includes both motional narrowing and many-body effects, and was evaluated using the best-fit parameters deduced in LSS. The solid straight line, which is a best fit to the data, would also result if the frequency-dependent term were attributed to g anisotropy under conditions of no motional narrowing. If the data of Fig. 1. for the 0.018-cm-thick sample were plotted they would fit on a parallel straight line.

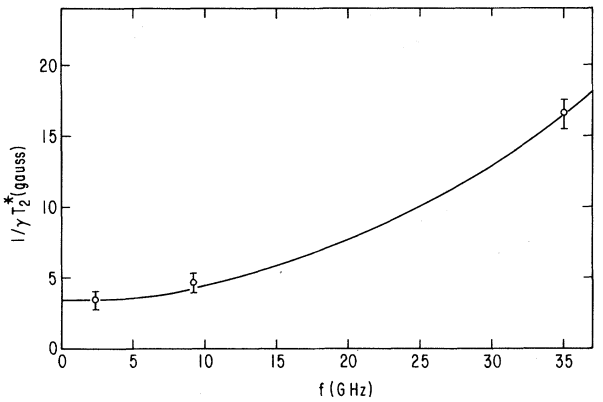

FIG. 3. Frequency dependence of the residual CESR linewidth, $\Delta H_R = 1/\gamma T_2^*$, for single-crystal Cu at 1.4 K. The experimental points are for a sample 0.005 cm thick. All the data were taken with samples cut perpendicular to the ⟨110⟩ axis, and the magnetic field parallel to that axis. The solid line is a parabola which indicates that ΔH_R may be expressed as the sum of a constant plus a frequency-dependent term proportional to ω^2. The quadratic dependence is interpreted as a consequence of g anisotropy in the extreme-motional-narrowing limit.

drawn in Fig. 2. Thus, we did not expect an appreciable further narrowing with reduced frequency. The residual linewidth data for the 0.004-cm sample of Fig. 1 are plotted in Fig. 2, and, as can be seen, appear to have a linear dependence upon the frequency, and deviate significantly from our predicted value at 1.27 GHz. (For comparison we also present the parabolic relationship that would result from fitting the 35- and 9.2-GHz residual linewidth data in the extreme-motional-narrowing limit.)

In contrast to the data for Al, the residual linewidth in Cu did not narrow appreciably, and does fit a parabolic dependence. In Fig. 3 we present ΔH_R for a 0.005-cm-thick Cu single crystal as a function of frequency. These data are consistent with the values of σ_g/g and B found in LSS.

The discrepancy presented in Fig. 2 might be resolved in several ways: (1) By choosing values for ΔH_R at the extremes of their errors, it is possible to obtain a fit to the FF theory. The problem with this approach is that the theory would then predict an appreciable narrowing of the 9.2-GHz linewidth between 10 and 20 K, which is not observed. In LSS we mentioned that there appeared to be some problems with the Al linewidths at higher temperatures, and if one wishes to interpret the discrepancy of Fig. 2 in this vein, we suggest that the assumption of a frequency-independent phonon contribution to the spin relaxation should be re-examined.[6] (2) The data in Fig. 2 may alternatively be taken to represent a true linear dependence of ΔH_R upon frequency. Holland and Dupree have made such a prediction when they suggested that Al CESR was to be interpreted in the non-motional-narrowing limit.[7] This interpretation would require a g-value scattering time several orders of magnitude longer than the resistivity scattering time, and could not explain the observed narrowing of the 35-GHz linewidth data above 15 K. (3) The FF theory, as in all other CESR theories known to us, results in expressions whereby the spin diffusion constant is proportional to the momentum scattering time, which in turn is taken to be isotropic. The electronic scattering in Al is known to be anisotropic from de Haas–van Alphen measurements.[8] Further, since a large-angle scattering [say from (100) to (110)] would not result in any change in

g value, whereas a small-angle scattering could very well be accompanied by appreciable changes in g values, the correct description of TESR may require a more complicated theory explicitly taking the anisotropy of both spin and momentum scattering into account.

In view of the foregoing results, we believe that a re-examination of the spin diffusion and relaxation processes in g anisotropic metals is warranted,[9] and particularly the consequences of incorporating such theory into the derivations for the TESR signals.

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[3]In Ref. 2 it is assumed that g is a scalar. The possibility that g is a tensor is treated in M. B. Walker, Phys. Rev. Lett. 33, 406 (1974).

[4]A detailed description of the 1.27-GHz spectrometer may be found in D. Lubzens, thesis, University of California, San Diego, 1975 (unpublished).

[5]The observed linewidth is corrected for finite thickness to yield $1/\gamma T_2^*$ as described by G. L. Dunifer, thesis, University of California, San Diego, 1968 (unpublished).

[6]Preliminary experiments with a doped Al sample are consistent with the assumption of a frequency-independent impurity contribution to the linewidth.

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[9]A detailed first-principles re-examination of the meaning of g values in anisotropic metals has been undertaken by W. Kohn and W. Lamb, University of California, San Diego, 1975 (unpublished).