# EFFECT OF MEAN FREE PATH ON THE RUDERMAN-KITTEL-KASUYA-YOSIDA SPIN-DENSITY OSCILLATIONS*

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Ruderman and Kittel[1] were the first to note that the response of an electron gas to a magnetic perturbation at the origin was nonmonotone and of the form

$$\sigma(r) \sim \frac{\cos 2k_F r}{r^3}$$

for large $r$, where $k_F$ is the Fermi wave number and $r$ is the distance to the point in question. Such spin-density oscillations are a general property of the electron gas[2,3] at metallic densities and arise whenever the conduction electrons scatter from a spin-dependent potential. One may view qualitatively the RKKY oscillations in a perfect metal as arising from an interference of the scattered outgoing spherical wave with the incident Bloch wave. If the metal is made nonperiodic, for example, by adding some nonmagnetic scattering centers, the exact states are no longer simple Bloch waves but linear combinations of many such waves with different $k$ values. This range of $k$ values leads to a damping of the interference oscillations in the manner familiar in all wave phenomena. In fact, one expects the spin-density oscillations in such a case to be of the form

$$\sigma(r) \sim \frac{\cos 2k_F r}{r^3} e^{-r/\lambda},$$

since the probability that the outgoing wave scattered from the impurity will reach a distance $r$ without undergoing a second scattering is $e^{-r/\lambda}$, where $\lambda$ is the mean free path.

The above qualitative idea has been dealt with from a more detailed theoretical point of view by de Gennes[4] and Mattis.[5] Mattis considered briefly the limit $k_F\lambda \ll 1$, a limit which is difficult to attain in a real physical system. De Gennes has treated the problem somewhat more generally, noting in particular that if the density of states at the Fermi level remains constant, there is a sum rule for the total susceptibility of the sample and the intuitive $e^{-r/\lambda}$ factor is only approximately correct. The major change, however, is the addition of a phase shift to the oscillations. The magnitude is found to be damped in approximately the expected manner.

In this paper we present a nuclear magnetic resonance (nmr) study of the spin-density oscillations in the vicinity of a Mn impurity in copper metal into which varying amounts of aluminum have been added as nonmagnetic scatterers. The experimental results clearly demonstrate the mean-free-path effect. The nmr technique is well suited for studying the spin polarization in metals; for a finite spin polarization gives rise, via the hyperfine interaction, to a local field as seen by the nucleus

$$H_{\mathrm{loc}}(r) = -\frac{A}{g_n \mu_n}\sigma(r),$$

where $A$ is the hyperfine coupling constant and $g_n$ and $\mu_n$ are the nuclear $g$ value and magneton, respectively. Since $\sigma(r)$ oscillates and diminishes in magnitude as $r$ increases, the dominant effect on the host-metal nmr is a broadening of the line[6] at low temperatures where the local-moment susceptibility is large. The temperature-dependent linewidth may therefore be used as a measure of the rms spin polarization in the host metal.

Figure 1 shows the experimental observation of the reduction of the spin-density oscillations

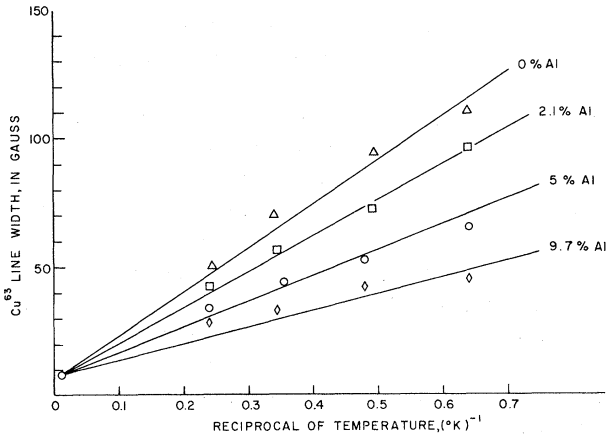


FIG. 1. Low-temperature linewidth of $Cu^{63}$ nmr at 8500 G in Cu-Mn alloys containing varying amounts of Al impurities. The Mn concentration is constant at 0.05 at.%.

as a result of adding aluminum scatterers to Cu-Mn alloys. The peak-to-peak derivative linewidth for the $Cu^{63}$ resonance is plotted versus reciprocal temperature. Although the linewidth measurement for such broad Lorentzian lines is limited to an accuracy of about 10%, the effect of the increasing Al content is clearly to decrease the slope. Susceptibility measurements on Cu-$Mn_xAl_y$ alloys[7] have demonstrated that the magnetic moment per Mn atom is unchanged with addition of aluminum-impurity content. Consequently, the slope of $\Delta H$ vs $1/T$ is a true measure of the conduction-electron polarization in the host at constant Mn concentration. The data indicate a strong reduction in polarization with decreasing mean free path (increasing concentration).

The samples indicated in Fig. 1 were prepared as a single batch and every attempt was made to keep the Mn content constant. First, a master Cu-Mn alloy was prepared by induction melting. The resulting ingot was divided equally into four pieces, and the four samples were remelted with the desired amount of Al included. All samples were then given a 72-h homogenizing anneal at 1000°C and were quenched into water. Quantitative analysis showed the Mn concentration to be constant to within experimental error (±0.005 at.%). Nmr studies of the $Al^{27}$ line showed no detectable Al out of solution (we estimate that 0.01% Al out of solution could be detected). The impurity concentrations used are all well within the known solubility limits for Mn and Al in copper, and the resulting alloys are expected to be single-phase solid solutions. Metallographic examination confirms this and shows no evidence of precipitation.

Figure 2 is a semilog plot of the slope $d(\Delta H)/d(1/T)$ for the various alloys as a function of aluminum concentration. Included are data from all samples, some of which were prepared completely independently of those in Fig. 1. Data from the different batches were normalized to the same Mn concentration as determined by chemical analysis. The various slopes are normalized to the pure Cu-Mn data as shown on the curve. The straight-line nature of Fig. 2 indicates

$$\langle\sigma(r)^2\rangle^{1/2} = \langle\sigma(r)^2\rangle_0^{1/2} e^{-\alpha c_1},$$

where $c_1$ is the concentration of aluminum impurities. Since $\lambda \sim (1/c_1)$, the rms polarization is found to vary experimentally as $\exp(-r_0/\lambda)$, where $r_0$ is a characteristic distance, in agreement with expectations.

Assuming a polarization of the damped RKKY form, the rms polarization may be calculated[8]:

$$\langle\sigma(r)^2\rangle^{1/2} = c_2\left[\frac{4\pi\langle\cos^2(2k_F r+\varphi)\rangle}{V}K^2\int_{r_c}^{R_c}\frac{e^{-2r/\lambda}}{r^6}r^2dr\right]^{1/2}.$$

The $\cos^2(2k_F r+\varphi)$ factor has been averaged using a random phase approximation, and the sum over all magnetic impurities yields the factor $c_2$, the concentration of Mn atoms. This should be valid so long as the Mn impurities are random and dilute. The integration in the averaging extends from an inner cutoff, $r_c$, to an outer cutoff, $R_c$. The inner cutoff must be included since the resonances from Cu sites very near a Mn are shifted by many kilogauss from the central $Cu^{63}$ line. Using the simple RKKY result with $J_{sd} \simeq 1$ eV as obtained from Behringer's[9] analysis of Cu-Mn nmr data, the nearest neighbor $Cu^{63}$ is shifted by $2\times10^4$ G. These separate satellites should not be included in the rms calculation since they clearly do not contribute to the linewidth. We choose as a cutoff, $r_c$, the distance at which the shift is reduced to be roughly within a measured

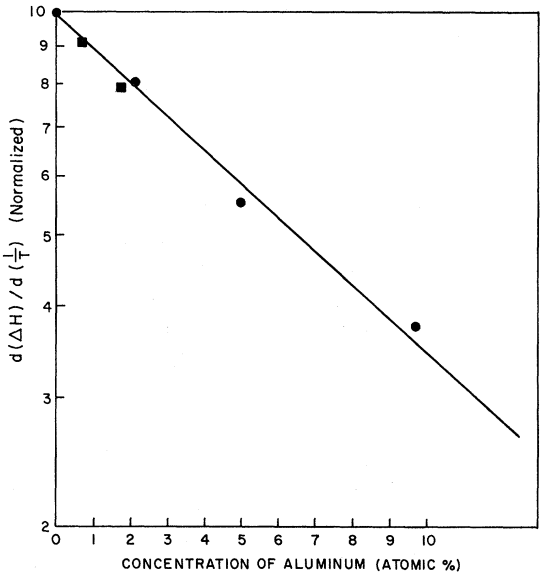


FIG. 2. Normalized slope $d(\Delta H)/d(1/T)$ for the Cu-Mn:Al alloys as a function of Al concentration. The circles represent data from Fig. 1. The squares represent data from samples of a separate batch.

linewidth; i.e., $\delta H(r_c) < 200$ G at the lowest temperatures. Using the above value for $J_{sd}$, the cutoff is determined as 5 lattice constants. The outer cutoff arises from the fact that for distances $r > R_c$, the shift becomes negligible compared with the homogeneous rigid-lattice width of 6 G for $Cu^{63}$. Again using Behringer's result, we find $R_c \simeq 20$ lattice constants. The mean free path, $\lambda$, may be estimated from measurements of the residual resistivity[10] of Al in Cu to be approximately 140 lattice constants at 1 at.% Al. Thus for concentrations less than 10% Al, $r_c/\lambda < 1$. Under these conditions the above expression is easily evaluated with the result that

$$\langle\sigma(r)^2\rangle^{1/2} \simeq \langle\sigma(r)^2\rangle_0^{1/2} e^{-r_c/\lambda},$$

where $\langle\sigma(r)^2\rangle_0^{1/2}$ denotes the rms polarization in the absence of impurities and is proportional to the local moment susceptibility.[9]

The result is in agreement with experiment as seen in Figs. 1 and 2. From the slope of the straight line of Fig. 2 we find $r_c/\lambda = 0.12$ at 1% Al. Finally, using the cutoff $r_c$ chosen above, we determine the mean free path from nmr data as 40 lattice constants at 1% Al. It is clear that the value for $\lambda$ obtained from resonance depends on the assumed value for $J_{sd}$. However, changing $J_{sd}$ by an order of magnitude only changes $r_c$, and thus $\lambda$, by a factor of 2, so the result is relatively insensitive to this choice. The apparent discrepancy in the resistivity and resonance values for $\lambda$ may be the result of the crude averaging procedure described above, and a more precise numerical evaluation is in progress. On the other hand, it is doubtful that the characteristic distance could differ from the above estimate by a factor of 2. The solution may simply be that the two experiments measure different mean free paths as a result of the $(1-\cos\theta)$ factor which diminishes the importance of low-angle scattering in resistivity theory.

Aside from affecting any phenomenon associated directly with the spin-density oscillations (e.g., nmr linewidths), the removal of such long-range effects would allow a study of the interaction of the conduction electrons with a single local moment. The study of this interaction, which leads to the well-known resistance minimum phenomenon,[11] is hampered experimentally[12] by the magnetic ordering resulting from overlap of the spin-density oscillations arising from different magnetic impurities. In fact, any study which attempts to investigate the single-impurity states is limited by this indirect exchange. Finally, the potential advantages of using paramagnetic alloys for adiabatic cooling is well known.[13] Again a decrease in the indirect exchange with a subsequent decrease in the "ordering" temperature would make such experiments feasible.

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[8]The factor $K$ in Eq. (5) is the proportionality constant and is given by (see Ref. 3)

$$K = -\frac{9\pi}{2} N^2 \frac{J_{sd}}{\epsilon_F} \langle S\rangle (2k_F)^{-3},$$

where $N$ is the number of conduction electrons per atom, $\epsilon_F$ is the Fermi energy, and $S$ is the local moment spin.

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