Investigation of Kondo Alloys and Compound by the Superconductive Proximity Effect

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The proximity effect between a superconductor and a normal metal is sensitive to the presence, amount, and state of order of the impurity spin in the normal metal. The present experiments were performed with $Cu_{0.99}Fe_{0.01}$, $Cu_{1-x}Mn_x$ ($x=0.0025$, 0.05), $Cu_{0.9975}Co_{0.0025}$ Kondo alloys, and with the Kondo compound $CeAl_2$. The alloys were sputtered first at temperatures ranging from about -100 to 530 °C followed by Pb sputtering at 77 °K. Electron diffraction showed that the Cu alloy films prepared at -100 °C were quasiamorphous and did not have the structure of bulk Cu. In such a state, regardless of the Kondo temperature of the bulk alloys, the magnetic impurities in the dilute alloys behave as random free spins; the spin depairing in the superconductive proximity effect is almost independent of the type of host metal (Cu or Mo) or the type of impurity (Fe, Mn, Co) and is proportional to the amount of impurity present. The $Cu_{0.95}Mn_{0.05}$ alloy, when deposited at -100 °C, becomes ferromagnetic at 5.5 °K and the magnetic transition is clearly exhibited by the proximity effect. Alloys prepared at and above room temperature have the bulk Cu structure. In the temperature range of the measurements (1–7.2 °K) which is well below the Kondo temperature of $Cu_{0.99}Fe_{0.01}$ ($\simeq 13$ °K as determined by resistivity), the $Cu_{0.99}Fe_{0.01}$ alloy films with the bulk Cu structure behave as an almost spinless system. The Cu-Mn alloys with the bulk Cu structure are antiferromagnetic at low temperature and give rise to a spin-depairing factor about 10 times smaller than the one corresponding to free random spins. The same qualitative result was obtained with Cr films, where the free-spin state can be achieved by sputtering at 77 °K. The proximity-effect experiments were also performed with the Kondo compound $CeAl_2$; above 3.5 °K $CeAl_2$ behaves as a spinless system which implies that $CeAl_2$ has a Kondo temperature above 7.2 °K. The antiferromagnetic transition at 3.5 °K is clearly shown by the proximity effect.

I. INTRODUCTION

The proximity effect of a superconducting film with various magnetic films (ferromagnets, antiferromagnets, dilute magnetic alloys) was investigated both by resistivity measurements[1] and tunneling experiments.[2,3] The transition temperature of Pb–magnetic-film sandwiches as a function of the Pb film thickness was fitted quite well[1] with a combined theory incorporating the de Gennes–Werthamer calculation of the proximity effect in nonmagnetic materials together with the Abrikosov-Gor'kov model of superconductivity in dilute magnetic alloys. As only two dilute magnetic alloys were studied[1] ($Mo_{0.99}Fe_{0.01}$ and $Pb_{0.971}Gd_{0.029}$), it was not directly established that the spin-depairing factor is proportional to the amount of spin present. Furthermore, the validity of the theory was not checked in very dilute alloys (impurity concentrations less than 1%) which actually is the concentration range where the theory should best apply. As the concentrated-ordered systems previously studied[1] were predominantly elements (Fe, Ni, Gd, Cr), it would be interesting to study the proximity effect on a concentrated-ordered alloy. The series of alloys investigated in this study provides a link between the dilute and concentrated systems and helps to confirm that the de Gennes–Werthamer–Abrikosov–Gor'kov[1] theory is valid over the whole concentration range.

It has been suggested[4] that Kondo alloys form below the Kondo temperature a quasibound state where the spin of the localized impurity is compensated for by the surrounding cloud of conduction-electron spin polarization. The existence of this compensated state was revealed experimentally by susceptibility[5,6] and Mössbauer[7] experiments. The proximity effect essentially probes magnetic spins with superconducting electron pairs over distances of the order of the coherence length. The proximity effect is therefore a convenient microscopic probe to study the compensation of localized spins below the Kondo temperature. One expects that proximity-effect measurements with Kondo alloys having a Kondo temperature much lower than the superconducting transition temperature will show the full random spin-flip scattering predicted by the lowest-order theory. On the other hand, when the Kondo temperature is sufficiently close to the transition temperature that higher-order corrections become important, or above the transition temperature, we might expect progressively less spin-flip scattering. Since the Kondo temperature is a rapidly varying function of the s-d exchange interaction, we expect to be able to sweep through these regions with relatively little change in the magnitude of the random spin-flip scattering. The ad-

vantage of the proximity effect over bulk transition-temperature depression studies in examining this effect is that we can measure Cu-based alloys whose magnetic and Kondo-effect properties have been thoroughly studied. Some of these alloys undergo magnetic ordering in intermediate concentration ranges, and it is possible to study the effect of this ordering on the spin-flip scattering by the proximity effect.

The bulk of the research on localized magnetic impurity states in metals was performed on well-annealed nearly perfect alloys.[8] Except for a proximity-effect study[1] on quasiamorphous $Mo_{0.99}Fe_{0.01}$ films deposited at 77 °K there is no experimental study on amorphous Kondo alloys. Films can be deposited in an almost amorphous state at low temperatures. The localized moments or Kondo state cannot be studied in such films by resistivity because the spin scattering is totally overshadowed by the boundary scattering of the small grains[9]; susceptibility measurements are equally difficult because of the small amount of material present in a film. The proximity effect seems to be the best method to study the Kondo state in these amorphous films. One can study with such experiments whether or not the Kondo temperature is affected by the degree of crystallinity of the matrix.

II. EXPERIMENT

All the films were deposited by the getter-sputtering technique in a previously described apparatus.[10] Films sputtered at room temperature and below were deposited on glass, while sapphire substrates were used for the higher deposition temperatures. In the sandwich preparation, the Cu alloy films, Cr, and $CeAl_2$ films were always deposited first; the Pb film was then sputtered at 77 °K and the sandwiches were kept at this temperature and measured without warming up. The Cu alloys comprising the sandwiches were kept at a fixed thickness (usually greater than 1000 Å) and were deposited at temperatures (T_D) ranging from − 100 to 530 °C. The 1250-Å Cr films used in this study were sputtered at 1100 °C from the same target which was used to deposit films at 77 °K in a previous study.[1] The $CeAl_2$ films were deposited at 800 °C and kept at a fixed 5000-Å thickness. The targets used for film deposition were made from arc-melted buttons[11]; the buttons were stirred and remolten many times to ensure homogeneity especially for the dilute Cu alloys. The composition of the dilute Cu-Mn alloys was checked by x-ray fluorescence analysis.[12]

The transition temperature of all the sandwiches is taken as the temperature where the first detectable normal voltage appears. The validity for this criterion has been demonstrated in numerous proximity effects[1,10] both in the nonmagnetic cases (Cu, Pt, Al) and in a dilute magnetic alloy $Pb_{0.971}Gd_{0.029}$ where experiments are fitted by the de Gennes–Werthamer theory[1] without any adjustable parameter. The width of most transitions is a few tenths of a degree. The normal residual resistivities of the component films of a sandwich are determined by sputtering separately each film on a substrate under the identical conditions used in the sandwich. Each film thickness is determined to within ± 5% by timing the duration of deposition, and then calibrating the deposition by weighing a thick film.

III. THEORETICAL ANALYSIS

The proximity effect between a superconducting and magnetic film is well described by a modification of the de Gennes–Werthamer theory.[1] The three equations which give an implicit solution for the transition temperature of the sandwich as a function of the thicknesses d_s and d_n of the superconducting and normal films, respectively, are[1]

$$\ln T_{cs}/T_c = \chi(\xi_s^2 k_s^2) , \tag{1}$$

$$\ln T_{cn}/T_c = \chi(-\xi_n^2 k_n^2 + \alpha/T_c) , \tag{2}$$

$$(\gamma \xi^2 k \tan kd)_s = (\gamma \xi^2 k \tanh kd)_n , \tag{3}$$

where $\chi(z) = \Psi(\frac{1}{2} + \frac{1}{2}z) - \Psi(\frac{1}{2})$ and Ψ is the digamma function. Here T_{cs}, T_{cn}, and T_c are the transition temperatures of the superconductor, the normal metal, and the sandwich, respectively; and ξ is the effective coherence length defined as

$$\xi = (\pi \hbar k_B \sigma / 6 T_c e^2 \gamma)^{1/2} , \tag{4}$$

where σ is the low-temperature conductivity and γ is the coefficient of normal electronic specific heat; and α, the spin-depairing factor, is defined as

$$\alpha = \hbar / 2\pi \tau_s k_B , \tag{5}$$

where τ_s is the electron spin-flip relaxation time. It should be pointed out that in Ref. 1, α was defined by the value given in relation (5) divided by T_{cs} which made it dimensionless. In the present study, α is a characteristic of the magnetic impurities and their normal matrix and is expressed directly in temperature units. For each alloy, α is determined by requiring Eqs. (1)–(3) to fit the experimentally determined plot of T_c vs d_s. As in the present experiments $d_n \gg \xi_n \geq k_n^{-1}$ and $T_{cn} = 0$ °K, Eqs. (2) and (3) reduce to the following equation:

$$(\gamma \xi^2 k \tan kd)_s = \gamma_n \xi_n (1 + \alpha/T_c)^{1/2} . \tag{6}$$

In all the figures shown in this paper, the curve corresponding to $\alpha = 0$ (no magnetic impurity) is represented by a solid curve. In all experiments performed in this study, $\rho_{Pb} = 1.5 \times 10^{-5}$ Ω cm and $\gamma_{Pb} = 1.72 \times 10^3$ erg/cm^3 °K^2.

The spin-flip scattering rate (or depairing parameter τ_s^{-1} was calculated for uniform superconducting magnetic alloys by Abrikosov and Gor'kov to lowest order in the s-d exchange energy.[13] Although this calculation pertains to a superconductor, it is just at the transition temperature and the conduction electrons are still normal. Therefore, the same calculation applies to the depairing strength of magnetic impurities in the present normal matrix. When the Kondo temperature T_K is small compared to T_c for the sandwich, this perturbation calculation should be valid, and τ_s should be temperature independent. Higher terms in perturbation theory for τ_s^{-1} will have logarithmic temperature dependences, however, as in Kondo's calculation of higher terms in the non-spin-flip scattering rate.[14] When these become comparable to the Abrikosov-Gor'kov term, perturbation theory breaks down.

The depression of T_c has been calculated by Zuckermann[15] using the self-consistent approximation devised by Nagaoka for normal metals.[4] The spin-flip scattering rate was also calculated by Suhl[16] from a set of assumptions which have been shown to be equivalent to the Nagaoka scheme. These theories predict that the depairing strength increases as T_c is lowered toward T_K, peaks at $T_c \sim 0.1 T_K$, and only drops below its high-temperature value for $T_c < 10^{-3} T_K$. This is a somewhat surprising result. As the spinless quasibound low-temperature state is formed, one might expect the depairing strength to decrease monotonically. We believe that our results show that this is in fact the case.

That the Nagaoka-Suhl approximation could be in such serious qualitative error in this prediction may be rationalized as follows: This approximation has been shown to predict both the asymptotic low-temperature behavior of the susceptibility and the nature of the approach to the low-temperature plateau of the resistivity incorrectly.[17] In Suhl's formulation, the scattering from the magnetic impurity is restricted to final (and virtual intermediate) states with just one electron or hole. It is clear that states with particle-hole pairs will have to be introduced to produce a theory which is in better agreement with the above experiments in the low-temperature limit. If such states are important, they must be included as additional "channels" in the scattering matrix. The requirement of unitarity will then cause the scattering in the one-electron channels to decrease. Since resistivity measurements confirm that the one-electron non-spin-flip channel has a large and increasing amplitude as the temperature is lowered through T_K, it is most probable that the amplitude is taken primarily from the one-electron spin-flip channel (the "depairing" channel).

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Cu-Fe Alloys

Cu-Fe alloys are Kondo alloys with a Kondo temperature (T_K) of approximately 14 °K.[5,8] In a proximity effect between a Cu-Fe alloy and a superconducting Pb film, the measurements are taken below the T_{cs} of Pb (7.2 °K) and therefore, in a temperature range where the quasibound state of the Cu-Fe alloy is well developed. Before interpreting any proximity effect, it is necessary to show whether or not the alloy films have the same property as the bulk alloys. Figure 1 shows the resistance as a function of temperature for a $Cu_{0.99}Fe_{0.01}$ film deposited at 800 °C; the resistance minimum which occurs in the same temperature range as in bulk alloys is a clear evidence of the Kondo effect. A plot of the resistance vs $\log_{10}T$ for the same film is shown in Fig. 2; choosing for T_K the midpoint of the linear portion of the log plot gives 13 °K in excellent agreement with bulk values. However, the value of the impurity contribution to resistance for this film ($\Delta\rho = \rho_{alloy} - \rho_{pure} = 1.4\ \mu\Omega$ cm) is 10 times lower than for bulk[5] (14 $\mu\Omega$ cm/%); the increase in resistivity from the minimum to the value obtained at the lowest-measured temperature is 0.14 $\mu\Omega$ cm which is also 10 times lower than for bulk.[5] These reductions are caused by the clustering of Fe in Cu; the solid solubility of Fe in Cu being extremely low (0.3 wt% at 800 °C), one can expect a large degree of clustering, especially in films, where the diffusion rates are high. By taking the ratio of the resistivity measurements on films to those obtained on bulk, one concludes that 0.1% Fe is free in a film prepared at 800 °C. As the temperature of deposition is decreased, the degree of clustering diminishes, thereby increasing the amount of free iron. At low deposition temperatures (– 100 °C), the resistance minimum can no longer be observed because the spin scattering

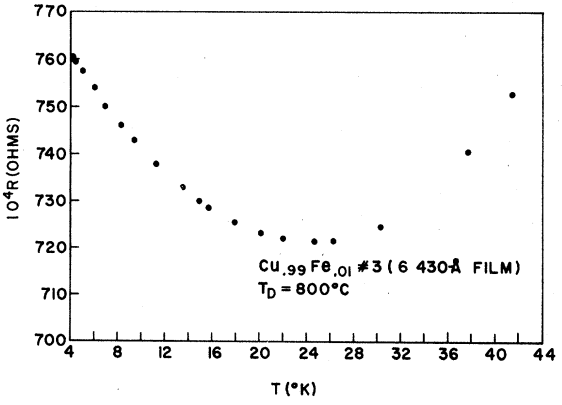

FIG. 1. Resistance as a function of temperature for a 6530-Å $Cu_{0.99}Fe_{0.01}$ film deposited at 800 °C.

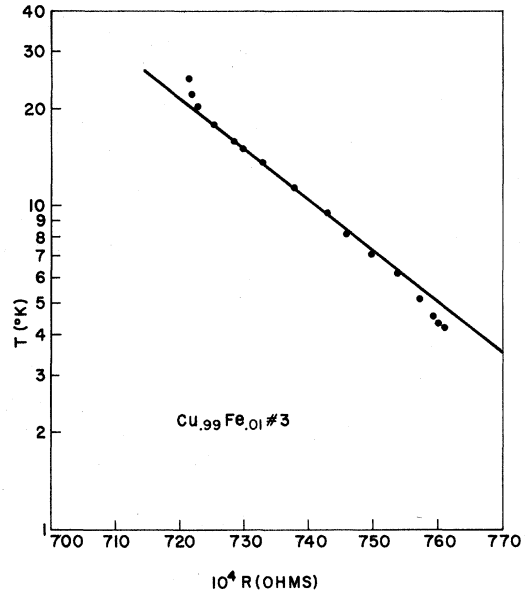

FIG. 2. Resistance vs $\log_{10}T$ for a 6430-Å $Cu_{0.99}Fe_{0.01}$ film deposited at 800 °C.

is totally overshadowed by the boundary scattering of the small grains. The lowest deposition temperature where the minimum can accurately be measured is 200 °C; at this temperature the resistivity increase from the minimum is 0.26 $\mu\Omega$ cm which after taking the ratio to the bulk value corresponds to 0.2% free iron. One can therefore conclude, that films prepared at temperatures below 200 °C will have at least 0.2% free iron.

While it is therefore desirable from the clustering point of view to deposit films at the lowest possible temperature, this may not be so from the structural point of view. Indeed, Cu and dilute Cu alloy films prepared at room temperature and above have the fcc structure and the lattice parameter of bulk Cu. This is not so for films prepared at − 100 °C. The electron diffraction taken at room temperature on such films is shown in Figs. 3(a) and 3(b). From the diffuseness of the diffraction rings, one can assert that such films are always quasiamorphous (grain size ranging from 10 to 30 Å). In one case, as shown in Fig. 3(a), the $Cu_{0.99}Fe_{0.01}$ film is bcc with a lattice parameter of 4.20 Å (±10%); in most cases as shown in Fig. 3(b), $Cu_{0.99}Fe_{0.01}$ films are fcc but with a lattice parameter of 4.28 Å as compared to 3.615 Å for bulk copper. Since in the proximity effects Cu alloy films are not warmed up as they are in the diffraction experiments, one can assume that the films in the proximity-effect experiments are at least as amorphous and distorted as shown by the electron diffractions. The resistivity of a $Cu_{0.99}Fe_{0.01}$ film prepared at − 100 °C (500 $\mu\Omega$ cm) does not change upon warming to room temperature implying that the bulk of the film is not changing. This does not preclude changes in the crystallinity of the surface which is precisely what both the electron diffraction and proximity effect measure.

The proximity effect with the various Cu alloys will be analyzed in terms of Eqs. (1) and (6); as the Cu alloys are dilute, the γ used in Eq. (4) will be that of pure copper[18] ($\gamma_{Cu} = 9.63\times10^2$ erg/cm^3 °K^2) while the resistivity will change with each deposition temperature and will be quoted in each respective figure. A problem which arises with the proximity effect is that the T_c of a sandwich may depend on the order of deposition of the films.[1,10] The lowest T_c (which yields the best proximity effect) corresponds[10] to Cu being deposited first, which will allow us to vary the deposition temperature of the Cu alloys while keeping the deposition of the Pb films fixed at 77 °K. The proximity effect of both Cu and $Cu_{0.99}Fe_{0.01}$ films deposited at 530 °C

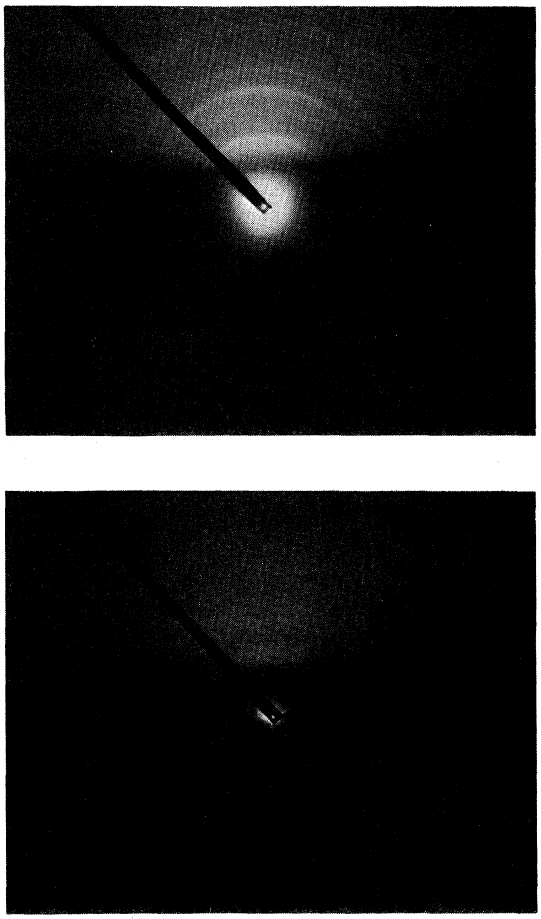

FIG. 3. (a) Electron diffraction taken at room temperature (RT) on a 1000-Å $Cu_{0.99}Fe_{0.01}$ film deposited at −100°C on glass (bcc film). (b) Electron diffraction taken at RT on a 1000-Å $Cu_{0.99}Fe_{0.01}$ film deposited at −100°C on glass (fcc film).

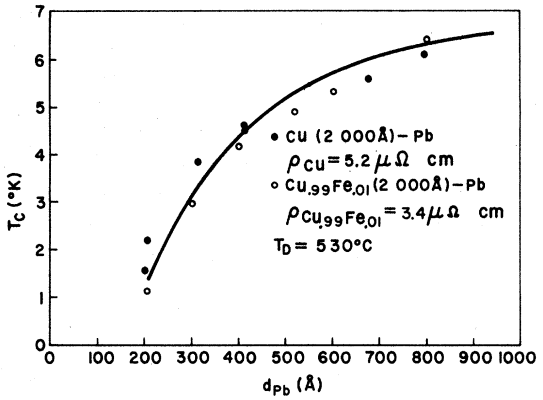

FIG. 4. Transition temperatures for Cu(T_D = 530 °C)-Pb and $Cu_{0.99}Fe_{0.01}$ (T_D = 530 °C)-Pb sandwiches as a function of the Pb film thickness.

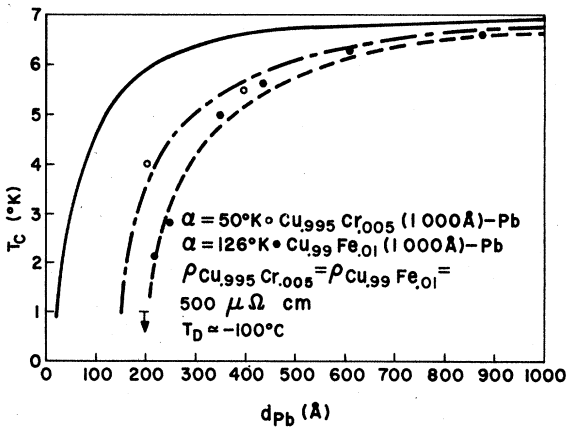

FIG. 5. Transition temperatures for $Cu_{0.995}Cr_{0.005}$ ($T_D \simeq -100$ °C)-Pb and $Cu_{0.99}Fe_{0.01}$ ($T_D = -100$ °C)-Pb sandwiches as a function of the Pb film thickness.

with Pb is shown in Fig. 4. The experimental points are fairly well averaged by the curve calculated from Eqs. (1) and (6) using $\alpha = 0$. Consequently, although at least 0.1% Fe is free in the $Cu_{0.99}Fe_{0.01}$ film, the Cu-Fe film behaves like Cu (i.e., in a spinless manner), or in other words, does not lead to any spin depairing.

The best way to make a proximity-effect measurement from an interface point of view is to deposit both films at 77 °K. Whenever the first film is deposited at some higher temperature, one may worry about condensing some impurities before deposition of the second film. This decoupling of the two films would lead to a higher transition temperature. One could therefore think that such an artifact is responsible for the $\alpha = 0$ fit found in the previous Cu-Fe experiments. This objection can be disposed of in three ways. First, Fig. 4 shows that Cu films prepared at the same temperature (530 °C) as the $Cu_{0.99}Fe_{0.01}$ films do not show any decoupling effect. Second, the proximity effect between Pt films, prepared at 1300 °K, and Pb were fitted exactly by the de Gennes–Werthamer theory.[10] Third, the absence of decoupling will be shown directly for the case of room-temperature deposition in the discussion on Cu-Mn alloys.

In contradistinction to the previous experiments are those where the $Cu_{0.99}Fe_{0.01}$ films are deposited at – 100 °C (Fig. 5). It is now apparent that the Fe has a strong spin-depairing effect; as a matter of fact, the value of $\alpha = 126$ °K used to fit the experiments is in very good agreement with a value of 148 °K previously[1] found for the proximity effect with another localized moment alloy $Mo_{0.99}Fe_{0.01}$.[19] This suggests that the value of the spin-depairing factor α is fairly independent of the nature of the matrix (Cu or Mo) as long, of course, as the impurity has an uncompensated local moment. Consequently, when the $Cu_{0.99}Fe_{0.01}$ film is deposited at – 100 °C, one finds that α has the full value expected for random spin-flip scattering by 1% Fe. This is not surprising, first of all because very little if any clustering is expected at such low deposition temperatures and second, as the Cu-Fe films do not have the structure of bulk Cu [Figs. 3(a) and 3(b)] one does not expect *a priori* these films to be in the compensated state. The Kondo temperature has been commonly defined as $T_K \simeq T_F e^{(-1/|J|\rho)}$, where T_F is the Fermi temperature, J is the exchange interaction, and ρ is the density of states per atom for one direction of spin evaluated at the Fermi surface. The observed stretching of the lattice parameter would lead to a larger value of ρ but to a smaller value of J (by reducing the overlap integral involved in the s-d mixing). The dominant effect seems to be the reduction of J and thus of T_K, so that the $Cu_{0.99}Fe_{0.01}$ films deposited at – 100 °C are no longer in the compensated state

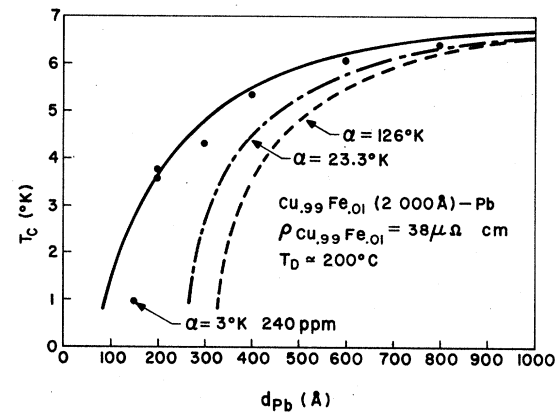

FIG. 6. Transition temperatures for $Cu_{0.99}Fe_{0.01}$ ($T_D \simeq 200$ °C)-Pb sandwiches as a function of the Pb film thickness – $\alpha = 23.3$ °K corresponds to 0.185% free iron.

above 1 °K. This explanation will find further support in the proximity effect with Cu-Co alloys. The proximity effect for two $Cu_{0.995}Cr_{0.005}$-Pb sandwiches is also shown in Fig. 5; the corresponding value of α of 50 °K is approximately half the value found for 1% Fe. It therefore seems that in the random spin-flip-scattering limit, α is proportional to the amount of impurity present irrespective of its nature (Cr or Fe); this point will be demonstrated with greater accuracy later on in the paper (Cu-Mn and Cu-Co alloys).

The Cu-Fe alloy films deposited at and above room temperature have the structure of bulk copper and are therefore well into the quasibound state below 7 °K. The proximity effect with $Cu_{0.99}Fe_{0.01}$ films deposited at 200 °C is shown in Fig. 6 where just as in the 530 °C case (Fig. 4) the data agree quite well with the curve $\alpha = 0$. The curve $\alpha = 126$ °K is shown for comparison; as one knows from resistivity measurements that at least 0.2% Fe is free, a curve labeled $\alpha = 23.3$ °K which corresponds to the random spin-flip scattering by that much iron is shown as well. The point labeled $\alpha = 3$ °K shows the biggest deviation from the curve $\alpha = 0$ and corresponds to a random spin-flip scattering by 240 ppm of Fe which is 9 times smaller than the actual concentration of free iron. The proximity effect with $Cu_{0.99}Fe_{0.01}$ films deposited at room temperature (RT) (Fig. 7) leads to the same conclusion. As the clustering at RT must be less than at 200 °C, the value of $\alpha = 6$ °K used to fit the data is much smaller than what one would expect from such a concentration of free spins. One experimental point in Fig. 6 ($d_{Pb} = 800$ Å) is very close to the curve $\alpha = 23.3$ °K. This is undoubtedly the result of scatter and of the uncertainty in the T_c measurement as the point is only 0.15 °K from the $\alpha = 0$ curve. This scatter does not affect the interpretation as the experimental points fall on the averaging curve ($\alpha = 0$ to $\alpha = 6$ °K) down to 1 °K where the quasibound state is fully formed as shown by resistivity measurements.[5] In conclusion, the proximity-effect data obtained on $Cu_{0.99}Fe_{0.01}$ films deposited at and above RT show that the spin depairing present in these films, even when clustering has been accounted for, is much smaller than what one would expect for random spin-flip scattering. In other words, in agreement with previous resistivity,[5] susceptibility,[6] and Mössbauer[7] experiments the quasibound state behaves as an almost spinless system. It should be noted that the temperature region of these measurements falls at the peak of the theoretical depairing strength vs T_c/T_K curve discussed previously.[15] That the depairing is greatly reduced instead is strong evidence that this theory is inadequate. On the other hand, the full random spin-flip scattering can be obtained in such alloys by low-temperature deposition which alters their crystalline structure and thereby, changes their T_K. One could argue that the difference between Cu-Fe films deposited at − 100 °C and those deposited above RT does not arise from a change of α but from a change in ξ caused by a change of γ. That this is not the case has been shown in two instances: Pt deposited at 77 and[1] 1300 °K and Cu deposited in this study at 530 °C (Fig. 1) and at[10] 77 °K and in both cases the change in ξ could be fully accounted for by the changes in σ.

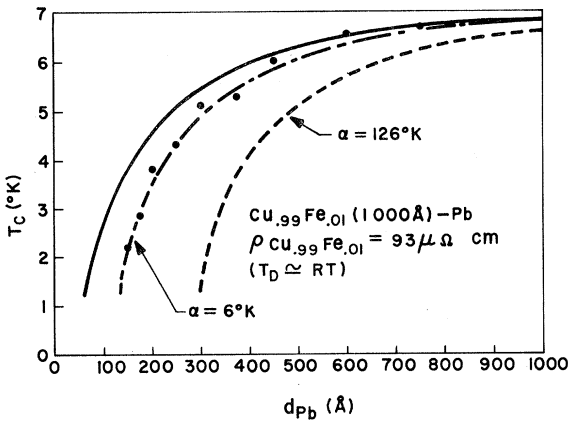

FIG. 7. Transition temperatures for $Cu_{0.99}Fe_{0.01}$ ($T_D \simeq$ RT)-Pb sandwiches as a function of the Pb film thickness.

It has been shown that the superconducting Kondo alloy La-Ce has a density of states which tends much more strongly to gaplessness for a given depression of the transition temperature than predicted by the Abrikosov-Gor'kov (AG) theory.[20,21] This aspect of the breakdown of the AG theory depends on many details of the coexistence of superconductivity and the Kondo quasibound state that do not enter the calculation of the depression of T_c, which depends only on α, the spin-flip-scattering rate in the normal state. Our result that this lifetime is lengthened in the Kondo state need not be inconsistent with increased gaplessness. In fact, one could interpret the results of Refs. 20 and 21 as indicating that a concentration of impurities producing a given degree of gaplessness is rendered less effective in depressing T_c by the Kondo effect.

B. Cu-Mn Alloys

Cu-Mn alloys have a low T_K (below 1 °K)[8] and therefore, will not be in the quasibound state in the range of the proximity-effect measurements. The solid solubility of Mn in Cu is very high (25% at RT) and therefore, even the most concentrated alloy used in this study (5 at. % Mn) should be free of the clustering problem encountered with the Cu-Fe alloys. Most experiments on Cu-Mn alloys have shown an antiferromagnetic behavior at low tem-

perature but the Néel temperature (T_N) varies depending on the method of measurement. The $Cu_{0.95}Mn_{0.05}$ alloy is certainly antiferromagnetic in the range of measurements (1–7.2 °K); Kouvel[22] who pointed out that Cu-Mn dilute alloys may be in a mixed ferromagnetic-antiferromagnetic state at low temperature reported from electrical-resistivity measurements a T_N of 150 °K for a 5.5-at.% Mn alloy. Recently, a value of 60 °K for a 4.3 at.% was obtained by electron-spin-resonance (ESR) measurements.[23] A general relationship $T_N = 2600c$ °K (where c is the concentration in at.%) which was obtained from specific-heat results[24] is in fair agreement with the above results. Applying this relationship to the most dilute alloy used in the present study ($Cu_{0.9975}Mn_{0.0025}$) yields a T_N of 6.5 °K. This result is in good agreement with the temperature of the resistivity maximum (T_{max}) observed by Schmitt and Jacobs[25]: They found $T_{max} = 5$ °K for a 0.2-at.% alloy and $T_{max} = 9$ °K for a 0.4-at.% alloy. One can therefore expect the $Cu_{0.9975}Mn_{0.0025}$ alloy to be antiferromagnetic in the whole range of the proximity-effect measurements.

Just as Cu-Fe alloy films deposited at −100 °C do not show the quasibound state of the bulk alloys, one can expect that Cu-Mn films deposited at −100 °C may not be antiferromagnetic as a result of the films not having the crystalline structure of bulk copper. Indeed, as shown in Fig. 8 the experimental points are extremely well fitted by Eqs. (1) and (6) using a value for α of 31.5 °K. This value of α for $Cu_{0.9975}Mn_{0.0025}$ is exactly $\frac{1}{4}$ of the α value used to fit the $Cu_{0.99}Fe_{0.01}$ proximity effect (Fig. 5). It is true that the points scatter from a value of $\alpha = 10$ °K to $\alpha = 55$ °K but this scatter should not only be attributed to α. Indeed as shown in Fig. 4 for pure Cu and in previous experiments,[1,10] a large amount of scatter comes from variations of the resistivity at the interface. Consequently, this selected value of α does not imply any great accuracy but simply shows that, given a certain scatter, $\frac{1}{4}$ of the value of α used for averaging the $Cu_{0.99}Fe_{0.01}$ data represents a fairly good averaging of the $Cu_{0.9975}Mn_{0.0025}$ data. This supports the statement made earlier that in the random spin-flip-scattering limit, the spin-depairing factor α is proportional to the amount of impurity present, irrespective of its nature (Cr, Fe, or Mn). In Ref. 26, de Gennes and Sarma have discussed the antiferromagnetic case and remarked that the spin-depairing factor α would be zero in that case. The fact that the value of α used in Fig. 8 is so close to the value expected for random spin-flip scattering shows, that very little, if any, antiferromagentic ordering is present in these films. The same kind of effect was already reported[1] for another antiferromagnetic case: Cr films when deposited at 77 °K were fitted with $\alpha = 150$ °K. This result as well as new Cr-Pb experiments will be discussed later on. Figure 8 shows three experimental points referred by crosses: These transition temperatures were obtained on sandwiches where the Cu-Mn film was warmed up from −100 °C to RT and then cooled to 77 °K before the Pb deposition. These experiments demonstrate that no decoupling occurs as a result of cooling from RT to 77 °K. This further supports the statement made earlier that the very reduced value of α observed in the proximity effect with Cu-Fe alloy films deposited at and above room temperature is not caused by a decoupling effect, but is the result of the quasibound state of the alloy. The experiments on the Cu-Mn films cycled to room temperature, far from showing a decoupling effect, actually show an enhanced proximity effect: The crosses are lower than the $\alpha = 31.5$ °K curve although the resistivity of such a cycled Cu-Mn film does not change upon warming up. This enhancement of the proximity effect can only be explained by a recrystallization of the surface which leads to a lower surface resistivity. As a consequence of this surface recrystallization, it is possible that Cu alloy films made at −100 °C and kept at 77 °K until measured are more amorphous and distorted than is shown by the electron diffraction of Figs. 3(a) and 3(b).

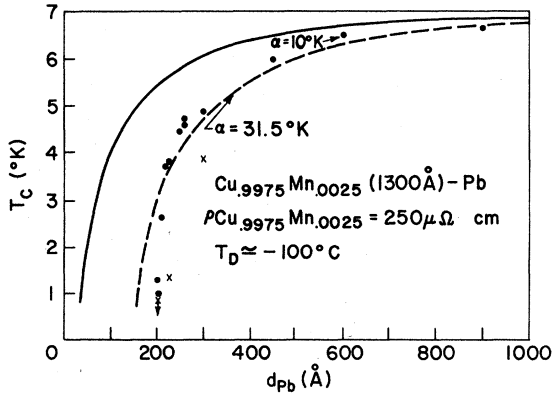

FIG. 8. Transition temperatures for $Cu_{0.9975}Mn_{0.0025}$ ($T_D \simeq -100$ °C)-Pb sandwiches as a function of the Pb film thickness. The crosses represent sandwiches in which the Cu-Mn film was warmed to RT and cooled again to 77 °K before the Pb depostion.

When the $Cu_{0.9975}Mn_{0.0025}$ alloy film is deposited at RT, it has the structure of bulk copper and one can expect the film to be antiferromagnetic below 6.5 °K. The proximity effect with such films is shown in Fig. 9. In agreement with the de Gennes–Sarma prediction,[26] one finds a greatly reduced value of α (3 °K). Comparing Figs. 8 and 9, one concludes that the antiferromagnetic ordering of the spins reduces the spin-depairing parameter α by a factor of 10. The same qualitative effect was observed on the elemental antiferromagnet

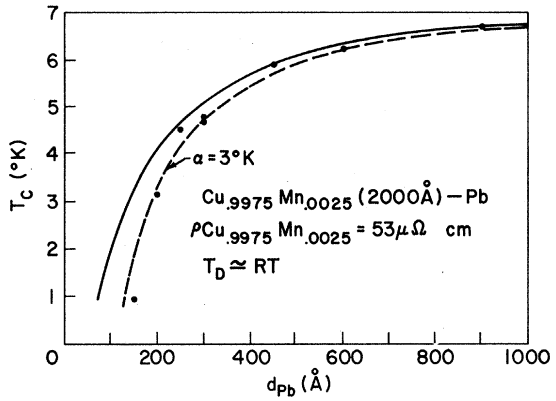

FIG. 9. Transition temperatures for $Cu_{0.9975}Mn_{0.0025}$ ($T_D \simeq$ RT)-Pb sandwiches as a function of the Pb film thickness.

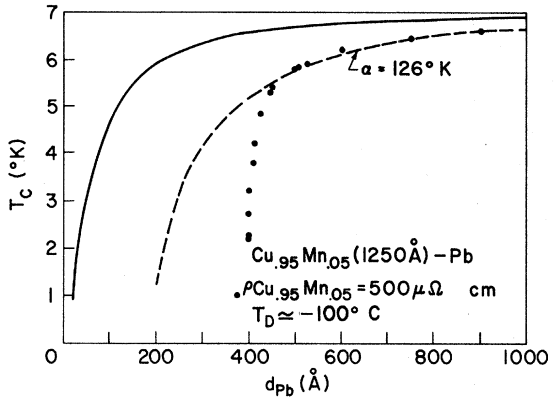

FIG. 11. Transition temperatures for $Cu_{0.95}Mn_{0.05}$ ($T_D \simeq -100$ °C)-Pb sandwiches as a function of the Pb film thickness.

Cr (see the discussion in Sec. IV D). The fact that α was not zero is most probably due to the fact that the film is not perfect and some spin depairing will occur at such defects as grain boundaries.

Let us now turn our attention to a concentrated alloy: $Cu_{0.95}Mn_{0.05}$. The result of the proximity effect with such an alloy film deposited at 200 °C is shown in Fig. 10. The value of α (15 °K) is quite low for such a high concentration of Mn as a result of the antiferromagnetic ordering. The proximity-effect curve for the same alloy deposited at −100 °C is shown in Fig. 11. The dashed curve which fits the experimental points with $\alpha = 126$ °K above 5.5 °K corresponds to an almost random spin-flip scattering. The reason that the spin-depairing factor in this case is not 20 times larger than in the $Cu_{0.9975}Mn_{0.0025}$ case (Fig. 8) is undoubtedly due to saturation effects arising from such high concentrations. Nevertheless, a comparison of Fig. 10 with the dashed curve of Fig. 11 still leads to the conclusion that the antiferromagnetic ordering of the spins reduces the spin-depairing parameter α by a factor of 8.

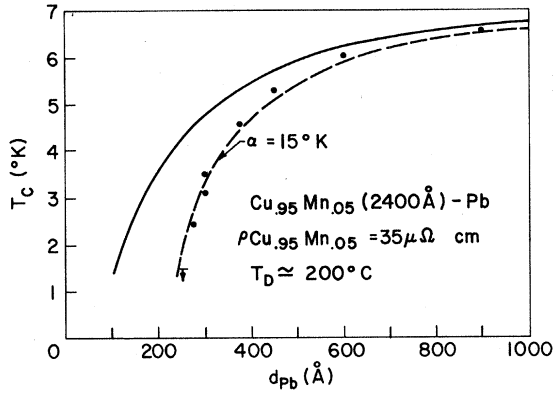

FIG. 10. Transition temperatures for $Cu_{0.95}Mn_{0.05}$ ($T_D \simeq 200$ °C)-Pb sandwiches as a function of the Pb film thickness.

The most striking feature of Fig. 11 is the very rapid drop in T_c below 5.5 °K over a very narrow range of Pb film thickness. The only phenomenon which could explain the enhanced proximity effect below 5.5 °K is ferromagnetism. It is true, however, that the spin-depairing factor (α) per impurity atom is largest for random spin-flip scattering; any ordered state will tend to decrease α. The enhanced proximity effect caused by the ferromagnetic transition can thus be explained in two ways. It is possible that while above 5.5 °K only the Mn atoms are contributing to the spin depairing, below 5.5 °K the Cu-Mn alloy becomes ferromagnetic as a whole thus behaving as a concentrated spin system.[1] On the other hand, when the Cu-Mn alloy becomes ferromagnetic, there appears an exchange field which will further depress the transition temperature of the superconducting Pb film.[27] The hypothesis of a ferromagnetic transition at 5.5 °K was checked in three different experiments. First of all, Table I shows that the transition temperature of a sandwich in the ferromagnetic region of the curve shown in Fig. 11 depends on the magnetic history of the sample. In Fig. 12, one sees that a sandwich in the paramagnetic region of the curve (above 5.5 °K) has the normal transition width of 0.3 °K for such sandwiches, on the other hand, sandwiches in the ferromagnetic region of the curve have a very dragged-out transition with a tail approach T_c. If one chooses in such a transition either the inflection point or a temperature 0.3 °K below the temperature where full normality has been restored, one obtains a temperature of approximately 5.15 °K. This is about what the transition temperature of the sandwich would be if the ferromagnetic transition did not take place. The

TABLE I. Transition temperatures of $Cu_{0.95}Mn_{0.05}$ ($T_D \simeq 100$ °C)-Pb sandwich ($d_{Pb} = 400$ Å) as a function of magnetic history.

Magnetic history	T_c (°K)
Sandwich as deposited (virgin)	2.71
After applying 17.4 kG parallel to film at 2.71 °K	2.55
After applying 17.4 kG perpendicular to film at 2.55 °K	2.47
After warming up to 77 °K and cooling through 5.5 °K with 17.4 kG perpendicular to film	2.43
After warming up to 77 °K and cooling in zero field	2.69

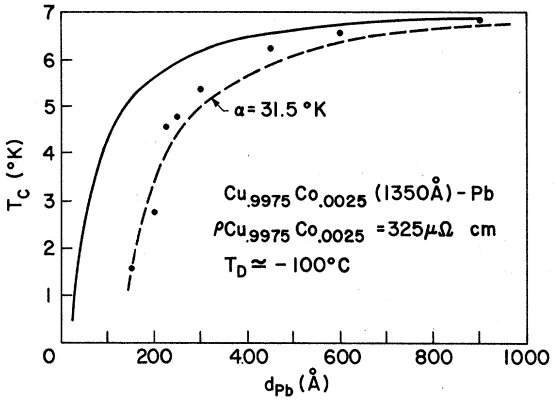

FIG. 13. Transition temperatures for $Cu_{0.9975}Co_{0.0025}$ ($T_D \simeq 100$ °C)-Pb sandwiches as a function of the Pb film thickness.

long tail is therefore a direct result of the ferromagnetic transition. Finally a 12 000-Å $Cu_{0.95}Mn_{0.05}$ film prepared at −100 °C and warmed up to RT was measured on a torque balance.[28] The warming up of the film should not influence the low-temperature state of the film as the resistivity of such a film remains unchanged. It was found that the film had magnetic hysteresis and a coercive force of approximately 3 kG at 1 °K. This clear evidence of ferromagnetism disappeared at 5.4 °K in excellent agreement with the proximity-effect measurements shown in Fig. 11.

C. Cu-Co Alloys

It was shown above, that while bulk Cu-Fe alloys have a T_K of 14 °K, T_K can be lowered below 1 °K by low-temperature deposition of these alloys. It would therefore be interesting to repeat such an experiment with another high-T_K alloy in order to demonstrate the generality of the effect and to possibly understand the mechanism responsible for the effect. This is the reason for choosing Cu-Co alloys where a T_K of 1000 °K has been reported.[29] According to a most recent theoretical analysis,[30] it may be better to say that Co does not form a well-defined local moment in Cu rather than to speak of such an elevated T_K. The proximity effect between Pb films and $Cu_{0.9975}Co_{0.0025}$ films deposited at −100 °C is shown in Fig. 13. The data can be fitted quite well with the value $\alpha = 31.5$ °K used for $Cu_{0.9975}Mn_{0.0025}$ in Fig. 8 which proves once again that the spin-depairing factor is simply proportional to the amount of spin present (Cr, Fe, Mn, or Co). In view of the scatter present, the proportionality is obviously not exact and certain impurities may depair more than others. However, as the average value of α for 1% Cr, Fe, Mn, or Co ranges from 80 to 150 °K, the difference between the scattering powers of these elements is certainly not large and certainly much smaller than the one reported for 4f impurities in La.[31] Furthermore, the proximity effect clearly shows that while Co has no moment in bulk Cu, it is magnetic in this stretched quasi-amorphous Cu obtained by low-temperature deposition. This result gives added support to the hypothesis given for Cu-Fe alloys that the reduction of T_K in these quasiamorphous alloy films is caused predominantly by a reduction of J.

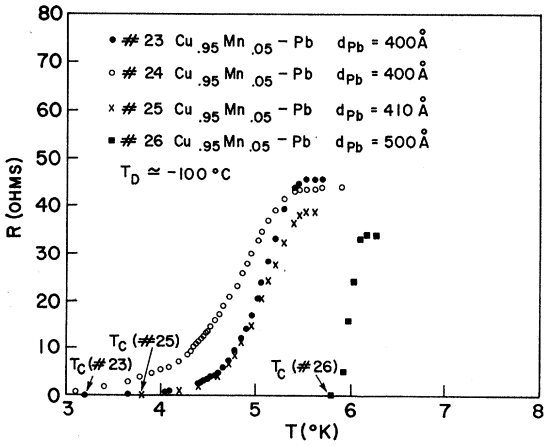

FIG. 12. Transition temperature width for four $Cu_{0.95}Mn_{0.05}$ ($T_D \simeq -100$ °C)-Pb sandwiches. The T_c's ($R = 0$) of these sandwiches are shown on Fig. 11.

D. Chromium

A previous proximity-effect study[1] showed that the transition temperatures of Pb-Cr sandwiches where Cr was getter sputtered at 77 °K could be fitted with $\alpha = 150$ °K. This result was in contradiction with de Gennes–Sarma's[26] theoretical prediction of $\alpha = 0$ for the antiferromagnetic case. This discrepancy was explained[7] by the fact that the theoretical argument applied only to a perfect bulk antiferromagnet; whereas in films sputtered

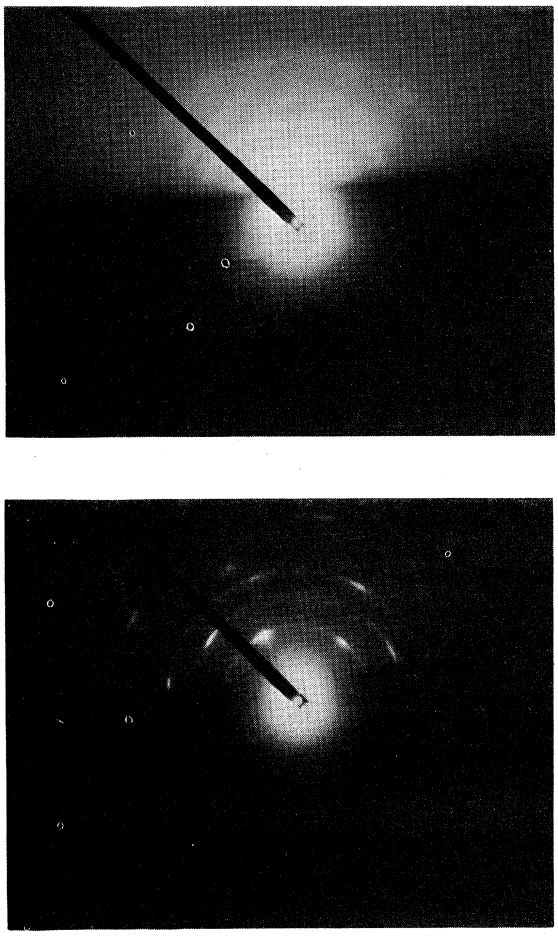

FIG. 14. (a) Electron diffraction taken at RT on a 300-Å Cr film deposited at 77 °K on glass. (b) Electron diffraction taken at RT on a 1200-Å Cr film deposited at 1100 °C on sapphire.

at 77 °K, the grain size was so small and spin scattering at domain walls sufficiently rapid that a random-impurity spin description was still valid. The validity of this explanation is verified by the fact that a Cr film sputtered at 77 °K with a resistivity of 370 $\mu\Omega$ cm is almost amorphous as shown by the extremely diffuse electron diffraction of Fig. 14(a). If on the other hand, one sputters Cr on a sapphire substrate held at 1100 °C, one obtains as shown in Fig. 14(b) a crystalline Cr film with preferred orientation and with the lattice parameter of bulk Cr. The Cr films deposited at 1100 °C have a resistance ratio of 2.3 and a low-temperature resistivity of 8.2 $\mu\Omega$ cm. The proximity effect with these films is shown in Fig. 15. It is now apparent, that as in the Cu-Mn case, the spin-depairing factor is almost zero in agreement with the de Gennes–Sarma prediction.[26] The small value $\alpha = 5$ °K is most probably caused by some remaining imperfections in the antiferromagnetic

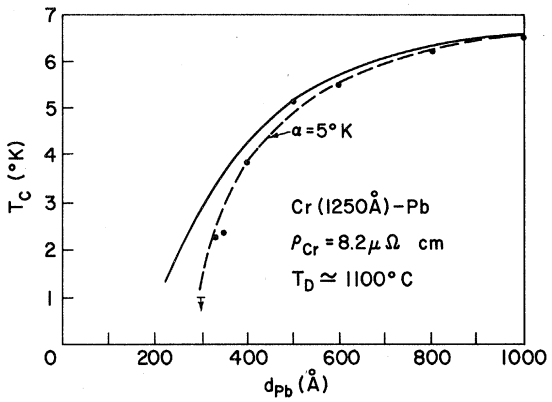

FIG. 15. Transition temperatures for Cr(T_D = 1100 °C)-Pb sandwiches as a function of the Pb film thickness.

ordering. One can therefore conclude, that in the proximity effect with an antiferromagnet, the spin-depairing factor is a measure of the degree of imperfection and of the number of defects.

E. $CeAl_2$

Recently, Buschow and Van Daal[32] have suggested the presence of the Kondo effect in the compound $CeAl_2$. Their resistivity data coupled with magnetic susceptibility suggest that $CeAl_2$ becomes antiferromagnetic below 6 °K. The pioneer work on these Laves-phase compounds suggested[33] that $CeAl_2$ was ferromagnetic below 8 °K. This result is in contradiction with the recent magnetic measurements of Swift and Wallace[34] who have shown a paramagnetic behavior at 4.2 °K and weak antiferromagnetism at 2.2 °K. In order to remove this ambiguity, it was decided to repeat the magnetic measurements on $CeAl_2$.[35] It is also noteworthy to point out that the magnetic measurements were taken on the $CeAl_2$ button from which the $CeAl_2$ films for the proximity effect were deposited. It is obvious from the susceptibility data shown in Fig. 16

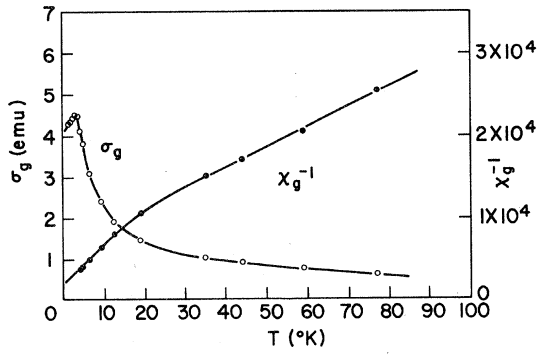

FIG. 16. Magnetic moment in 15 300 Oe and inverse susceptibility measured as a function of temperature on the $CeAl_2$ target used for film deposition (Ref. 32).

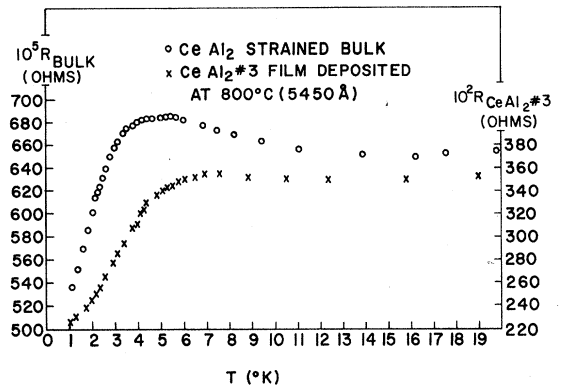

FIG. 17. Resistance as a function of temperature for strained $CeAl_2$ bulk and for a 5450-Å $CeAl_2$ film deposited at 800 °C on sapphire.

that $CeAl_2$ becomes antiferromagnetic at 3.5 °K. This result is not only in good agreement with the previous observations of Swift and Wallace,[34] but also with recent heat-capacity measurements by Hill and da Silva[36] who clearly show a magnetic transition at 3.4 °K. A piece was cut from the $CeAl_2$ button used in the magnetic measurements, and resistance measurements taken on this strained sample are shown in Fig. 17. The resistance as a function of temperature is also shown for a $CeAl_2$ film deposited on sapphire at 800 °C. The data shown in Fig. 17 for the strained bulk and the film are in good qualitative agreement with Buschow's data[32]: There is a shallow resistance minimum around 16 °K and a rapid decrease of the resistance below 6 °K. Although these resistivity data do not exactly agree either with specific-heat measurements[36] or with susceptibility measurements (Fig. 16), it is clear from Fig. 17 that the $CeAl_2$ films are representative of the bulk properties. Most Kondo alloys at low temperatures show a negative transverse or longitudinal magnetoresistance.[25,37] The transverse magnetoresistivity of $CeAl_2$ measured at 1 °K is negative and linear in H^2 up to 10 kG and increases faster than H^2 above 10 kG; the residual resistivity decreases by 4.6% in a field of 17.4 kG.

The proximity-effect measurements on $CeAl_2$-Pb sandwiches are shown in Fig. 18. The theoretical interpretration of these results in terms of Eqs. (1) and (6) is hampered by the fact that γ is not known for $CeAl_2$. It is, as a matter of fact, doubtful whether one will ever be able to extract a value for γ from specific-heat measurements: At low temperatures, as shown by Hill and da Silva[36] the specific heat is dominated by the antiferromagnetic transition; at higher temperatures it will be extremely difficult to subtract the lattice term because of the contribution to the specific heat by the Kondo effect. Consequently, the curve shown in Fig. 18 is the result of fitting the data above 3.5 °K using Eqs. (1) and (6) with $\alpha = 0$ and the measured resistivity of $CeAl_2$. The value of $\gamma = 7.73 \times 10^3$ erg/°K^2 cm^3 results from such a fit. The right-hand side of Eq. (6) suggests that one could fit the experimental points above 3.5 °K with $\alpha \neq 0$ and a smaller γ value. If one fits the data above 3.5 °K using for γ the smallest possible value, 1.46×10^3 erg/°K^2 cm^3 which is the value for pure Al, one obtains the maximum possible value for α of 22 °K. The curve $\alpha = 22$ °K agrees almost as well with these data as the curve $\alpha = 0$. It is, however, unreasonable to expect γ for $CeAl_2$ to be as low as for pure aluminum, and as a result α, if not zero, is certainly much lower than 20 °K. For such a concentrated system as $CeAl_2$ (see $Cu_{0.95}Mn_{0.05}$) a value of α of even 20 °K indicates a pair-breaking parameter much reduced from the random spin-flip value.

The most striking feature shown by the data of Fig. 18 is the sharp departure from the curve at 3.5 °K in excellent agreement with the antiferromagnetic transition observed by specific-heat[36] and susceptibility measurements (Fig. 16). As it is, obviously, the localized spins on the Ce atoms which order antiferromagnetically at 3.5 °K, one is again faced with the puzzling fact that above 3.5 °K this dense spin system behaves in a spinless manner. This fact can be explained if one assumes that just like Cu-Fe alloys, $CeAl_2$ is a Kondo compound with a $T_K \geq 7.2$ °K. In this case, below 7.2 °K $CeAl_2$ is in a quasibound state and will therefore behave in a proximity effect as a spinless material ($\alpha = 0$). Below 3.5 °K, the antiferromagnetic transition breaks up the compensation and in agreement with the results obtained with Cu-Mn alloys, gives rise to a larger value of α.

The high value of T_K inferred from the proximity effect can be verified from the susceptibility measurements shown in Figs. 16. The curve labeled

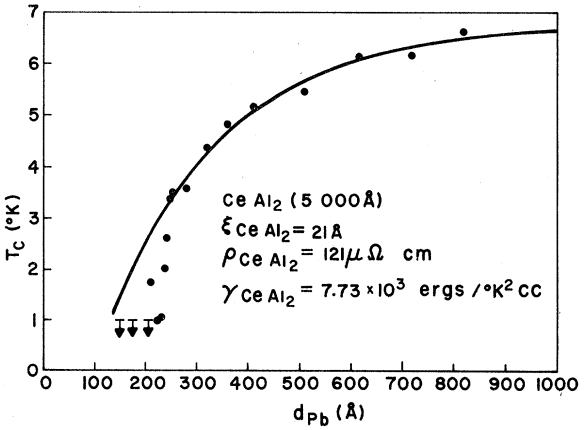

FIG. 18. Transition temperatures for $CeAl_2$(T_D = 800 °C)-Pb film thickness.

χ^{-1} is very similar to the one obtained on Cu-Fe alloys.[8] It has been widely suggested that

$$\chi^{-1}(T=0)=3kT_K/\mu_e^2 . \quad (7)$$

On the other hand, as in the case of Cu-Fe the range above $3T_K$ can be approximated by the Curie-Weiss law[8]

$$\chi^{-1}=(3k/\mu_e^2)(T+4T_K) . \quad (8)$$

One can therefore determine T_K from an extrapolation of the high-temperature data (above 20 °K) setting $\chi^{-1}=0$ in relation (8) which yields $T_K=6.25$ °K. It also follows from relations (7) and (8) that $T_K=\chi^{-1}(T=0)/(\partial\chi^{-1}/\partial T)_{T>20\,^\circ\mathrm{K}}$ which incorporates the low-temperature data extrapolated through the antiferromagnetic transition and yields $T_K=7.2$ °K.

V. SUMMARY AND FUTURE WORK

The proximity effect has proven to be a very useful tool in the study of Kondo alloys and compound in their various magnetic states. In dilute alloys, the spin-depairing factor was found to be proportional to the amount of impurity, irrespective of the nature of the matrix (Cu or Mo) or of the impurity (Cr, Fe, Co, Mn) as long, of course, as the impurity is magnetized in that matrix. The concentrated alloys show saturation effects and spin-depairing factors quite similar to the ones previously found in elemental concentrated spin systems. Except for ferromagnetism which because of its exchange field shows more spin depairing than the random spin-flip scattering, all other ordered states show a greatly reduced spin depairing. In agreement with previous susceptibility and Mössbauer measurements, the quasibound state appears as a spinless state. The antiferromagnetic state shows a very reduced spin depairing which approaches zero with the perfection of the antiferromagnetic order. Alloy films prepared at low temperatures have a very different magnetic behavior from the more perfect bulklike films prepared at higher temperatures. The most striking example was a quasiamorphous $Cu_{0.95}Mn_{0.05}$ film with a 5.5 °K Curie temperature. It is planned to further study such amorphous alloys with susceptibility measurements. The proximity-effect study of the $CeAl_2$ compound leads to the conclusion that $CeAl_2$ is a Kondo compound with $T_K \geq 7.2$ °K and is therefore in a quasibound state down to 3.5 °K where the compensated state is broken up by the antiferromagnetic transition. It is also interesting to point out that the proximity effect gave a value of 7.7×10^3 erg/°K^2 cm^3 for the γ of $CeAl_2$ which one may not be able to obtain from direct specific-heat measurements.

Recently, Kaiser and Zuckermann[38] using McMillan's[39] tunneling model for the proximity effect have calculated the excitation spectrum in normal superconducting sandwiches for the case where the normal film contains magnetic impurities. This calculation was made in the clean limit. Preliminary tunneling experiments were made on $Cu_{1-x}Mn_x$ – Pb sandwiches ($x=2500$, 1000, and 200 ppm) and $Cu_{0.99}Fe_{0.01}$ – Pb sandwiches. In order to satisfy the clean limit, the copper alloys are deposited at 200 °C on the aluminum oxide, followed by the usual deposition of Pb at 77 °K. The sandwiches were measured at 1 °K without warming and except for an increasing degree of gapless superconductivity with impurity concentration and with alloy film thickness,[40–43] no anomalous peak in the density of states as reported by Mihalisin *et al.*[44] on Au-Fe alloys was observed.

ACKNOWLEDGMENT

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correspond to a value of α as defined by relation (5) in the text of 77 °K. If one instead fits to the glass-$Mo_{0.99}Fe_{0.01}$-Pb data, one obtains the value of 148 °K quoted in the text.

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Interaction of 9.3-GHz Longitudinal Phonons with Electrons in Superconducting Films

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Previous studies of the interaction of longitudinal phonons with superconducting Al-Pb junctions have been extended to include nonuniform strains, arising when the thicknesses of the metal films are not negligible compared to the wavelength of the sound and/or when the wave front makes a small angle θ with the plane of the junction. It is found that nonuniform strains produce an electric field proportional to the strain gradient inside the "local" superconducting Al film. This result confirms the theories of Dessler *et al.*, Herring, and Harrison who calculated the gravity-induced electric fields. The strain-induced field in the aluminum excites electromagnetic waves in the junction which are enhanced due to the traveling-wave interaction in the strip line formed by the junction. The electromagnetic field gives rise to an extra tunneling current which exhibits resonant behavior as a function of the angle θ. In the "nonlocal" Pb films, the interaction of the electrons with the strain wave is described by absorption and emission of phonons, and the extra tunneling current due to this process is essentially independent of θ. This behavior enables us to separate the extra current due to the Al film and that due to the Pb film. The agreement between theory and experiment is found satisfactory.

I. INTRODUCTION

It was shown several years ago that the interaction between sound waves and electrons in metals can be observed directly by measuring the phonon-induced current in superconducting tunnel junctions.[1-3] In the case of compressional waves,[1,3] this extra current was attributed to a relative motion of the Fermi energies in the two metals due to the compressional strain. In the interpretation of the above experiments it was assumed that the strain was spatially uniform. The present work,

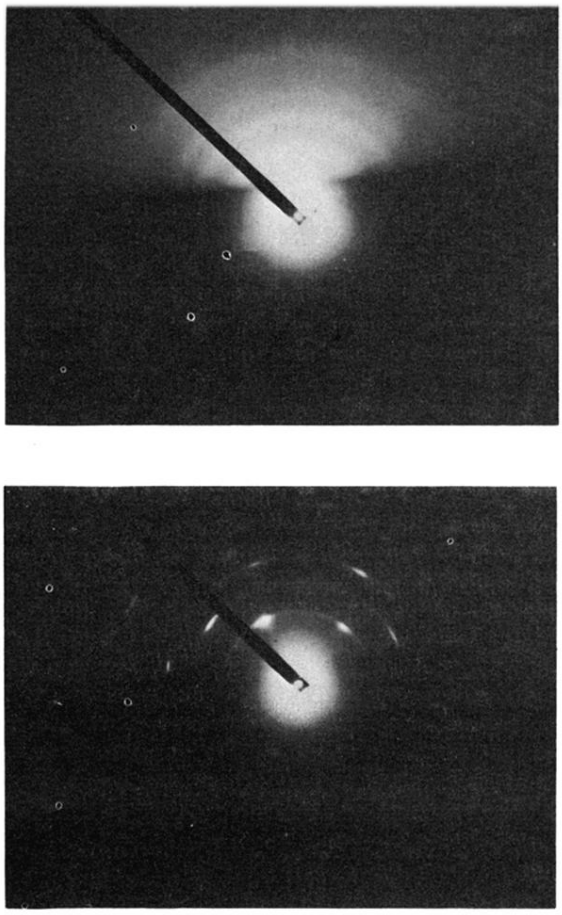

FIG. 14. (a) Electron diffraction taken at RT on a 300-Å Cr film deposited at 77 °K on glass. (b) Electron diffraction taken at RT on a 1200-Å Cr film deposited at 1100 °C on sapphire.

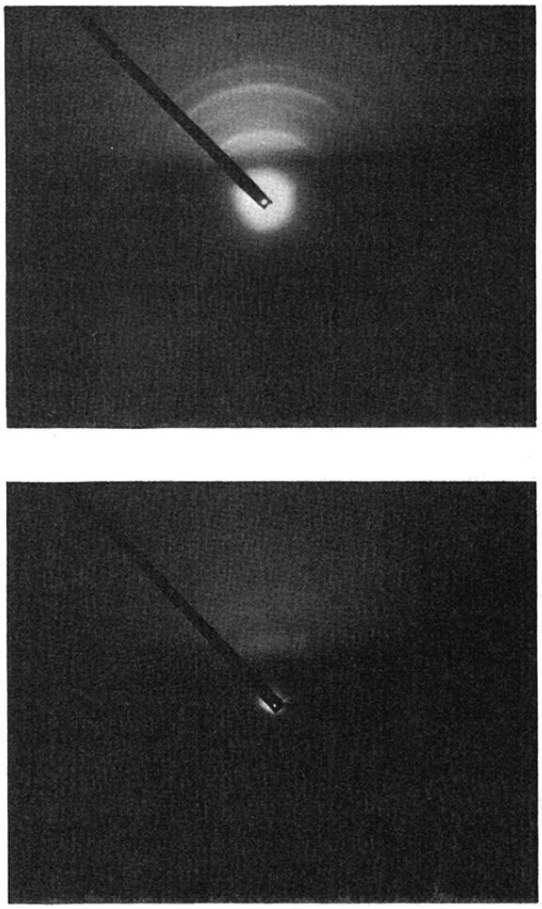

FIG. 3. (a) Electron diffraction taken at room temperature (RT) on a 1000-Å $Cu_{0.99}Fe_{0.01}$ film deposited at −100°C on glass (bcc film). (b) Electron diffraction taken at RT on a 1000-Å $Cu_{0.99}Fe_{0.01}$ film deposited at −100°C on glass (fcc film).