Magnetic Proximity Effect

J. J. HAUSER
Bell Telephone Laboratories, Murray Hill, New Jersey 07974
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The penetration depth of the spin polarization of a magnetic metal (Fe, Ni, or Cr) into a nonmagnetic metal or alloy with a high exchange-enhanced spin susceptibility (Pd and $Ni_{0.58}Rh_{0.42}$) was studied by use of the superconducting proximity effect. Pb-Pd-(Fe or Cr) and Pb-$Ni_{0.58}Rh_{0.42}$-Ni sandwiches as well as Pb-Mo-Cr were prepared by the getter-evaporating and getter-sputtering techniques at 77°K and their transition temperatures were measured without warming up. Since the magnetic penetration depth is very small, the Mo sandwiches were used to rule out the possibility of pinholes through the nonmagnetic metal. Also, the Mo sandwiches permit the evaluation of the superconducting proximity effect in the Pd sandwiches, since molybdenum has a low spin susceptibility, but at the same time the penetration of superconducting pairs is approximately the same in Mo and Pd. The ranges of polarization of Fe and Cr into Pd were found to be approximately 20±10 Å, and the range is possibly less for Ni into $Ni_{0.58}Rh_{0.42}$. Finally, Ni_3Ga-Pb sandwiches (Ni_3Ga was chosen because of its very high spin susceptibility) had transition temperatures consistent with Ni_3Ga being a normal nonmagnetic compound, from which it is concluded that if spin fluctuations are present in Ni_3Ga, they do not lead to appreciable spin depairing.

I. INTRODUCTION

THE large magnetic moments associated with first-row transition-metal atoms dissolved in a nonmagnetic metal with high exchange-enhanced spin susceptibility, such as palladium, have been extensively studied. Experimentally, giant moments have been reported[1] for Fe in Pd, and the distribution of magnetic moment has been investigated by neutron scattering.[2] Recently, weak ferromagnetism was discovered in Pd-Cr alloys.[3] Theoretically, Clogston[4] studied the range of a static spin polarization in Pd and pointed out that the range of the spin polarization of Pd conduction electrons by Fe is about 10 Å. In general, the high exchange-enhanced spin susceptibility in the nearly ferromagnetic materials such as Pd, Ni-Rh alloys, and Ni_3Ga has been explained in terms of the spin-fluctuation theory.[5,6] It would therefore be of interest to measure directly the range of the spin polarization in a metal such as palladium; this can be accomplished by depositing a magnetic film (such as Fe, Ni, or Cr) on a thin film of Pd. The magnetic proximity effect of the magnetic film on Pd can be measured by studying the superconducting proximity effect of this magnetic film–Pd sandwich on a superconducting Pb film. Alternatively, one may try to reveal the spin fluctuations directly through the depairing effect on a superconducting thin film (Ni_3Ga-Pb sandwiches), since spin fluctuations are damped spin waves, and since it is well known that a magnetic spin leads to strong depairing in a superconducting metal.[7]

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II. EXPERIMENTAL PROCEDURE

The Pb-Pd-(Fe or Cr) sandwiches as well as Pb-Mo-Cr and Pb-$Ni_{0.58}Rh_{0.42}$-Ni were deposited on glass substrates by the getter-sputtering[8] and getter-evaporating[9] techniques at 77°K. More precisely, the Pb, Pd, Mo, and $Ni_{0.58}Rh_{0.42}$ films were getter-sputtered in argon at 2 W, while the Fe, Cr, and Ni films were getter-evaporated from a tungsten heater. The $Ni_{0.58}Rh_{0.42}$ sputtering target was rolled from a bulk piece annealed *in vacuo* at 1100°C.[10] The Ni_3Ga-Pb sandwiches were deposited on sapphire substrates. The Ni_3Ga sputtering target was made from an arc-melted button annealed *in vacuo* for a week at 1000°C.[11] The Ni_3Ga films were made by sputtering at 15 W, with a substrate temperature ranging from 650 to 1000°C; the Ni_3Ga films were then cooled to 77°K with the shutter over while sputtering is proceeding and, finally the Pb film is sputtered over. In all cases, the sandwiches were mounted on the holder under liquid nitrogen and measured without warming up in order to avoid spurious diffusion effects. In each class of sandwiches, the lead film was kept at a constant thickness: 300 Å for Pb-Pd-(Fe or Cr) and Pb-Mo-Cr, 375 Å for Pb-$Ni_{0.58}Rh_{0.42}$-Ni, and 320 Å for Ni_3Ga-Pb. The Pd, Mo, and $Ni_{0.58}Rh_{0.42}$ films ranged in thickness from 10 to 60 Å, and the Ni_3Ga films were kept at constant thickness of 6900 Å. The Fe, Cr, and Ni films were approximately 150 Å thick.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Since one is trying to appraise the magnitude of the magnetic proximity effect in the Pb-Pd- or Mo-Cr-type

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[10] I am indebted to E. Bucher for this alloy.
[11] I am indebted to D. D. Dorsi for the Ni_3Ga compound as well as for the Mo target.

TABLE I. Physical properties of Pd and Mo films on glass.

	Pd $\rho(3720\text{ Å})=1.14\times10^{-4}\,\Omega$ cm, $\gamma=1.06\times10^{4}$ erg/°K^2 cm^3, $\xi=18.5$ Å				Mo $\rho(4300\text{ Å})=4.1\times10^{-4}\,\Omega$ cm, $\gamma=2.35\times10^{3}$ erg/°K^2 cm^3, $\xi=20.7$ Å		
d (Å) sputtering time	d (Å) light transmission	R (Ω)	R_{calc}	d (Å) sputtering time	d (Å) light transmission	R (Ω)	R_{calc}
10	16	3×10^{7}(RT)	2×10^{3}	10	7	6.6×10^{4}(RT)	7.5×10^{3}
14	14	5.6×10^{3}	1.43×10^{3}	15	18	3.7×10^{4}	5×10^{3}
20	17.5	2.34×10^{3}	10^{3}	20	16.5	6.9×10^{3}	3.75×10^{3}

sandwiches, two important facts should be kept in mind. First, any normal film such as Pd or Mo (which has a low spin susceptibility), when thin enough, becomes superconducting in contact with a superconductor and will therefore lead to a strong superconducting proximity effect of the Pb-Cr type.[7] Furthermore, since the spin polarization range is expected to be very small,[4] the normal films (Pd, $Ni_{0.58}Rh_{0.42}$) will have to be thin and could contain some pinholes, which again would directly lead to a Pb-Cr proximity effect. In other words, in order to measure the depth of penetration of the spin polarization from Cr into Pd, it is necessary to verify that the Pd film is free of pinholes (or at least that the concentration of pinholes is such that the mean distance between pinholes is much larger than the lead coherence distance $\simeq$100 Å) and to estimate the superconducting proximity effect taking place in the Pb-Pd sandwich.

The properties of thin Mo and Pd films are compared in Table I. Since palladium has a large spin susceptibility and, thus, a large coefficient of normal electronic specific heat γ, molybdenum was chosen because of the low conductivity σ of films deposited at 77°K, so that ξ, which is proportional to $(\sigma/\gamma)^{1/2}$, is approximately the same for Mo and Pd. Since ξ is a measure of the penetration distance of superconducting pairs into the normal metal, this means that the superconducting proximity effect of Pb is about the same on Mo and Pd. Consequently, the superconducting proximity effect being about the same in Pb-Pd-(Cr or Fe) and in Pb-Mo-Cr sandwiches,[12] any difference in their transition temperature can be ascribed to the penetration of the spin polarization into the palladium. As shown in Table I, Mo and Pd films become continuous at approximately the same thickness (10 Å). Furthermore, when the thickness reaches 20 Å, the resistance of the films is only twice the calculated one. The resistance was evaluated using bulk resistivity (i.e., the resistivity of a 4000-Å film); if, instead, one used a resistivity corrected for surface scattering with a mean free path of 30 Å, the calculated resistance would be 60% higher and would almost agree with the experimental one. This agreement between calculated and experimental resistance implies that 20-Å Mo and Pd films are quite uniform in thickness and that only a small fractional area can be covered with pinholes. The similar physical properties of Mo and Pd films imply that the probability of pinhole formation must be the same for both films. Therefore, if a Pb-Pd-Cr sandwich has a lower transition temperature than a Pb-Mo-Cr sandwich (with same Pb thickness and the Pd thickness of the first sandwich being equal to the Mo thickness of the second), it is difficult to argue that it is because of the stronger proximity effect of Pd, or because of a larger propensity for pinhole formation in Pd.

The transition temperatures of Pb-Pd-(Cr or Fe) and Pb-Mo-Cr sandwiches as a function of the Pd and Mo film thickness are listed in Table II. The proximity effect between a superconducting film and a normal magnetic or nonmagnetic film is described by the equations[7]

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

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

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

where the effective coherence distance ξ is defined as

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

and

$$\alpha=\hbar/2\pi\tau_sk_BT_{cs}\,. \tag{5}$$

Here T_{cs}, T_{cn}, and T_c are the transition temperatures of the superconductor, the normal metal, and the sandwich, respectively, d_s and d_n are, respectively, the thickness of the superconducting and normal films, σ is the low-temperature conductivity, γ is the coefficient of normal electronic specific heat, and τ_s is the electron relaxation time. The parameter k_s^{-1} is a measure of the penetration distance of normal electrons in the superconductor, while k_n^{-1} measures the penetration distance of superconducting pairs into the normal film. It follows from Eq. (2) that when $T_{cn}=0$°K and $\alpha=0$, $k_n^{-1}=\xi_n$.

When the Pd or Mo film thickness becomes very large, the effect of Cr or Fe is no longer felt and we are left with the simple superconductor–normal metal

[12] The molybdenum films used in this study (ranging in thickness between 10 and 4300 Å), as well as those reported in Ref. 7, were found not to be superconducting above 1.05°K. Furthermore, a Pb(80-Å)–Mo(300-Å) sandwich was not superconducting above 1.05°K, thus showing that Mo films deposited on lead have the same superconducting properties as those deposited on glass.

TABLE II. Transition temperatures of Pb-Pd-(Cr or Fe) and Pb-Mo-Cr sandwiches as a function of the Mo and Pd film thickness.

d_{Pd} or d_{Mo} (Å) sputtering time	T_c(Pd-Fe) (°K)	T_c(Pd-Cr) (°K)	T_c(Mo-Cr) (°K)
0	<1	1–2.7; 1.6	1–2.7; 1.6
10	1.5	...	...
15	...	...	4.8
20	2.1, 2.5	1.6, 1.8, 2.2, 2.6	5, 5.4, 5.4
30	1.9, 2.4	3.4, 3.9	...
40	3.1	4.9	...
50	3.7	...	...
60	4.2	...	...
∞	4.8 (theoret., 4.2)	4.8 (theoret., 4.2)	6.2 (theoret., 5.9)

proximity effect, which can be calculated using Eqs. (1)–(3) with $\alpha=0$ and $T_{cn}=0$°K. This calculation yields 4.8°K for the Pb-Pd sandwiches, and 5.9°K for the Pb-Mo. The experimental values shown in Table II are somewhat higher; this results from the fact that the experimental values were measured on glass-Pb-Pd and glass-Pb-Mo sandwiches, while it is the reverse order (glass-Pd-Pb and glass-Mo-Pb) that yields the lowest transition temperatures.[13]

As already measured before,[7] and as verified in this study, the transition temperature for a glass-Pb-Cr sandwich with a 300-Å Pb film thickness varied between 1.0 and 2.7°K. This experimental value can be calculated[7] from Eqs. (1)–(3), using $T_{cn}=0$°K and $\alpha=21$. This wide scatter comes from the steep dependence of T_c on lead film thickness especially in the vicinity of 300 Å. It is obvious from Table II that a Pb-Pd-Cr sandwich with 20 Å of Pd has the same transition temperature as a Pb-Cr sandwich, while a Pb-Mo-Cr sandwich with a 20-Å film of Mo still has a transition temperature very close to a Pb-Mo sandwich. As a matter of fact, even when the Mo thickness is reduced to 15 Å, the transition temperature remains quite high. Consequently, one can conclude that the 20 Å film of Pd has been completely permeated by the spin polarization of the Cr or, in other words, that 20 Å of Pd in contact with Cr behaves like chromium.

TABLE III. Physical properties of $Ni_{0.58}Rh_{0.42}$ films and transition temperatures of Pb-$Ni_{0.58}Rh_{0.42}$-Ni sandwiches as a function of the $Ni_{0.58}Rh_{0.42}$ film thickness.

$\rho(2470\ \text{Å})=3.7\times10^{-5}\ \Omega$ cm
$\gamma=1.5\times10^{4}$ erg/°K² cm³

d (Å) sputtering time	d (Å) light transmission	$R(\Omega)$	R_{calc}
20	21	5.7×10^{4} (RT)	338
40	40	196	169

$d_{Ni_{0.58}Rh_{0.42}}$ (Å)	T_c(Pb-$Ni_{0.58}Rh_{0.42}$-Ni) (°K)
0	<1
15	4
20	3.6
30	4.3
40	4.3
∞	4.6, 4.5 (theoret., 4.3)

The same type of experiment can be performed by replacing Cr by Fe. Fe has a stronger proximity effect than Cr, as shown by the transition temperature of a Pb-Fe film (<1°K). The Pb-Fe experiments were fitted[7] with Eqs. (1)–(3), using $T_{cn}=0$°K and $\alpha=14$. The transition temperatures of Pb-Pd-Fe sandwiches, as a function of the Pd film thickness, show an arrest point at about 30 Å of Pd. This implies that the spin-polarization wave from the iron has penetrated the 30 Å of Pd. If the Pd thickness is reduced to 10 Å, we then obtain a transition temperature intermediate between that of a Pb-Fe sandwich and that of a Pb-Pd(20 Å)-Fe sandwich; since 10 Å is the onset of continuity for Pd films, it is quite possible that this transition temperature is the result of pinholes through the Pd film. The slightly different behavior of Pb-Pd-Fe sandwiches from that of Pb-Pd-Cr can be understood if one assumes that once the Pd film becomes permeated by the spin-polarization wave of the iron, it still is not identical to iron. In other words, although the 20- or 30-Å Pd film in contact with Fe is "magnetic," the α to be used in Eqs. (1)–(3) for this "magnetic" Pd film is not necessarily equal to the α for Fe ($\alpha_{Fe}=14$); it is coincidental that the α pertinent to the Pd film in contact with Cr was identical to $\alpha_{Cr}=21$. We can therefore conclude that the penetration depth of the spin polarization of Fe or Cr into Pd is of the order of 20 ± 10 Å.[14] The uncertainty of 10 Å comes mainly from the first 10 Å of the film, which is known to be the onset of continuity on glass (see Table I). However, since the 20-Å film

[13] The cause of this reversal has been fully discussed in Refs. 7 and 8. One could think of measuring glass-Pd-Pd sandwiches, which is the proper order in that case, but then one would be forced to measure glass-Cr or Fe-Pd-Pb sandwiches, which, in the limit of thin Pd films, tend to glass-Cr-or Fe-Pb sandwiches, which is the wrong order for these sandwiches as shown in Ref. 7.

[14] This result should not be confused with the much longer range of polarization reported by previous authors: J. C. Bruyère, G. Clerc, O. Massenet, R. Montmory, L. Néel, D. Paccard, and A. Yelon, J. Appl. Phys. **36**, 944 (1965); O. Massenet, IEEE Trans. Magnetics **4**, 26 (1968). These authors obtained their spurious results on films deposited at room temperatures, in which appreciable diffusion had taken place.

TABLE IV. Physical properties of Ni_3Ga films and transition temperatures of Ni_3Ga-Pb sandwiches $\gamma Ni_3Ga = 1.29\times10^4$ erg/°K² cm³.

Temp. of deposition (°C)	ρ(4.2°K) (Ω cm)	ρ(RT)/ρ(4.2°K)	ξ (Å)	$T_{c\,\mathrm{expt}}$ (°K)	$T_{c\,\mathrm{theoret}}$ (°K)
1000	3.2×10^{-5}	1.5	32	4.2	3.5
800	1.5×10^{-5}	3	47	3.5	3.1
800	9.8×10^{-6}	4	59	2.8	2.9
650	$3\ \times10^{-5}$	2.2	33	3.6	3.5

has almost the calculated resistance, and since the onset of continuity should be lower on Pb than on glass, one can expect the reported film thicknesses to be quite accurate.

It would be interesting to repeat the same experiments replacing Pd by a $Ni_{0.85}Rh_{0.42}$ alloy, the idea being that since $Ni_{0.58}Rh_{0.42}$ has about four times the Stoner susceptibility enhancement factor of Pd,[15] it may, accordingly, have a larger spin-polarization wave penetration depth. Since this did not turn out to be the case, it was again necessary to use ultrathin films of $Ni_{0.58}Rh_{0.42}$ in the triple sandwiches. However, in this case, it was impossible to make a reliable film thinner than 20 Å. The data are shown in Table III and with the present scatter, the only conclusion which can be drawn is that the penetration depth of the nickel spin polarization is less than 15 Å and, therefore, smaller than in Pd. This is in agreement with susceptibility measurements on $Ni_{0.58}Rh_{0.42}$ alloys doped with iron.[16]

In the case of Ni_3Ga, it is impossible to perform the previous experiments as Ni_3Ga must be sputtered at high temperatures in order to obtain the proper crystallographic structure. One can, however, hope to exhibit the spin fluctuations in Ni_3Ga by studying the transition temperatures of Ni_3Ga-Pb sandwiches. Indeed, if spin fluctuations are present in Ni_3Ga, this spin depairing should result in a lower transition temperature than if Ni_3Ga were simply a normal metal; in other words, the transition temperature of such sandwiches should correspond to a nonzero value of α in Eq. (2). However, the calculated transition temperatures from Eqs. (1)–(3), using $T_{cn}=0$°K and $\alpha=0$, agree very well with the experimental values as shown in Table IV. It should be pointed out that the best resistivity ratio of 4 obtained in the Ni_3Ga films is appreciably lower than the bulk value of 50.[17] Bearing this in mind, these experiments show that, if spin fluctuations are present in Ni_3Ga, they do not lead to any spin depairing. This can be explained by the fact that the relaxation time for spin fluctuations is of the order of 10^{-14} sec, which is short compared to the mean lifetime of a superconducting pair. It is interesting to point out that there are no nickel clusters in the Ni_3Ga films, since such clusters would lead to a very strong spin depairing and, thus, to very low transition temperature.[7]

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