PHYSICAL REVIEW B VOLUME 11, NUMBER 1 1 JANUARY 1975

Low-temperature resistivity of Yb in Au, Ag, and Au-Ag alloys

Y. Talmor and J. Sierro
Département de Physique de la Matière Condensée, 32, Boulevard d'Yvoy, CH-1211 Genève 4, Switzerland

(Received 15 April 1974)

We present resistivity measurements on $(\mathrm{Au}_x\mathrm{Ag}_{100-x})_{100-y}\mathrm{Yb}_y$, which cover the concentration range between $x = 0$ and 100 at.%, and $y = 0$ and 2 at.%. The resistivity minima observed are found to be a combination of two mechanisms, which can be separated only by a suitable heat treatment. The first effect is due to the Yb behaving as a solid solution and is explained in terms of the shift of the Fermi level as the host's composition is varied. The second effect results form the presence of intermetallic phases inside the matrix, essentially YbAu_3. Of these two mechanismes, that due to the intermetallic phase dominates. The resistivity of some intermetallic compounds YbAu_x has also been measured and found to show resistivity minima, larger than any of those observed in the (AuAg)Yb system.

I. INTRODUCTION

It has been shown, from measurements of lattice parameters of *Au*Yb alloys[1] and from the electrical-resistivity[2] and magnetic-susceptibility[3] measurements in *Ag*Yb, that Yb is divalent in Ag and trivalent in Au. These facts lead to the assumption of a variable valence for Yb in $\mathrm{Au}_x\mathrm{Ag}_{100-x}$ alloys.

Boes *et al.*,[4] who were the first to measure the resistivity of this system, found a minimum on the silver-rich side. Allali *et al.*[5] have since studied the susceptibility and the thermoelectric power (TEP) of certain alloys of the system. The anomalous Curie-Weiss temperatures which they observed in the concentration range between 5 and 30% gold, have been attributed to the Kondo effect, in agreement with the results of Boes *et al.* The TEP measurements give the critical concentration of the host at which the Kondo effect appears; it is situated at about 7% gold.

Recent observations of the EPR spectra[6] and the Mössbauer effect[7] of ytterbium in gold, on the other hand, make the gold-rich side seem particularly interesting.

Previously, the only direct evidence of the Kondo effect in these alloys emerged in the early measurements of Boes *et al.*

We present here resistivity measurements which cover the concentration range from pure gold to pure silver.

The metallurgy of the ternary system *AuAg*Yb is relatively troublesome. This is due on the one hand to the great difference of the solubility of ytterbium in gold and silver, and on the other hand to its great affinity to form intermetallic compounds with the components of the host. This fact makes it essential to know the metallurgical state of the alloy for any interpretation of the experimental results.

We have chosen to measure the resistivity at low temperatures because this kind of measurement provides direct evidence of the Kondo effect, and it permits a qualitative control of the metallurgy of the system.

This paper is divided into three principal parts. After a review of the experimental procedure, we give in Sec. III A the results obtained for ytterbium dissolved in gold and silver separately. The system $(\mathrm{Au}_x\mathrm{Ag}_{100-x})_{99.5}\mathrm{Yb}_{0.5}$ is studied in Secs. III B and C, while Sec. IV is also devoted to this system but there the alloys are less dilute and also contain several intermetallic compounds of ytterbium with gold.

II. EXPERIMENTAL PROCEDURE

The two components of the host alloy were provided by Engelhard and each has a purity of 99.999%. The ytterbium, provided by the Nuclear Corp. of America, has a purity of 99.9%. The alloys described in Sec. III A were prepared with triply distilled high-purity ytterbium provided by Bucher and Schmidt from the Bell Telephone Laboratories.

All the alloys, including the intermetallic compounds, were prepared in an arc furnace using a copper crucible and a tungsten electrode. The host alloy was first prepared, then a part of it was remelted together with the required amount of Yb. The two alloys were subsequently given the same thermal treatments and samples from them were measured simultaneously. The ingots were drawn into wires of 0.1 mm in diameter through diamond dies. The resistance measurements were always made on wires at least 1 m long. For certain measurements, the samples were given one or both of the following heat treatments: (i) Immediately after fabrication a 7-day homogenization was given at 560 °C in a sealed quartz tube containing argon; (ii) immediately after drawing into wire a 1-h anneal was given at either 560 or 850 °C in an argon atmosphere before oil quenching to 0 °C.

The ytterbium content was analyzed for some

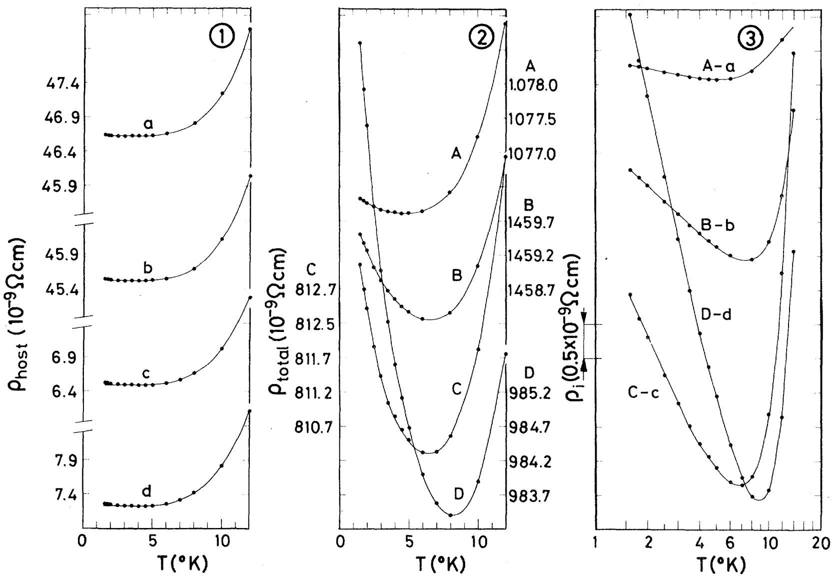

FIG. 1. Temperature dependence of the electrical resistivity of $\text{Au}_{99.5}\text{Yb}_{0.5}$ (nominal concentration) for different heat treatments. Curve 2 shows the behavior found for the alloys; Curve 1 that for the hosts alone; and Curve 3 that for the impurity. (These curves are the difference between Curves 2 and 1 in each case.) Throughout, only relative values of $\rho_i(T)$ are discussed because of the uncertainty in the determination of the size factor for a sample. The heat treatments were (A, a) as-cast; (B, b) 1 week at 560 °C before coled working; (C, c) 1 h at 560 °C after coled working; and (D, d) 1 week at 560 °C before coled working followed by 1 h at 560 °C after coled working. All samples containing Yb were prepared using triply distilled high-purity Yb.

samples while for the rest the concentrations indicated are the nominal values. Where it seemed necessary we also took x-ray pictures of powdered samples as a qualitative check of the metallurgical state.

The resistivity was measured by a dc four-probe method using a Honeywell Rubicon resistance bridge and a Keithley 147 null detector. The experimental error in the wire's size factor does not exceed 5%.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Ytterbium dissolved in silver and gold

Previous results suggest that ytterbium acts as a divalent nonmagnetic impurity in silver. On the other hand, ytterbium is trivalent in gold; its $4f$ level can be split and the solute ion can become magnetic. The Kondo effect is impossible in the first case but probable in the second.

We have made two series of measurements of *Au*Yb and *Ag*Yb alloys. The nominal amount of ytterbium in both cases was 0.5 at.%. One sample of each series was analyzed by x-ray fluorescence with the following results: 0.32 ± 0.02 at.% for the gold-based alloys and 0.28 ± 0.02 at.% for the silver-based ones. Both series had identical thermal treatments of which details are given in the captions to Figs. 1 and 2. The contribution $\rho_i(T)$ of the impurity is deduced from Matthiessen's rule:

$$\rho_{\text{tot}}(T)=\rho_{\text{host}}(T)+\rho_i(T).$$

In the case of *Au*Yb (Fig. 1), the resistance minimum indicates a magnetic behavior for the ytterbium, as expected. Murani,[8] who recently measured the resistance of several rare-earth metals dissolved in gold, observed the same behavior for an alloy analogous to case B of Fig. 1. The influence of the thermal treatment is seen to be extremely important. The resistance minimum is weakest for the as-cast sample (A), is still more pronounced in the homogenized sample (B), is still more pronounced in the sample annealed after coled working (C), and reaches its greatest value in the sample which has been homogenized before coled working and then annealed (D). The logarithmic temperature dependence is seen to exist up to 5 °K.

Table I is a summary of the different parameters which can be deduced from our resistivity measurements: $\Delta\rho_{\text{min}}=\rho^{\text{tot}}_{1.5\,^\circ\text{K}}-\rho^{\text{tot}}_{\text{min}}$; T_{min} determined from the curves of $\rho_{\text{tot}}(T)$; and $\Delta\rho_i/\Delta(\ln T)$ determined from the curves of $\rho_i(T)$. From these values we have calculated J_{sf}, using Kondo's formula (corrected by the de Gennes factor):

$$\rho_i(T)=c\rho_0[1+2J_{sf}(g-1)\eta(\epsilon_F)\ln T],$$

with

$$\rho_0=\frac{\pi m\eta(\epsilon_F)}{e^2\hbar n}(g-1)^2 j(j+1)J_{sf}^2.$$

Here n is the number of conduction electrons per unit volume and c is the impurity concentration. The other symbols have their usual meanings. The

TABLE I. Summary of some parameters deduced from the present resistivity measurements for *Au*Yb and *Ag*Yb alloys. The citations relating to the heat treatments are the same as those in Figs. 1 and 2.

Alloy		$-\Delta\rho_i/\Delta(\ln T)$ ($10^{-9}\Omega$ cm)	$-\frac{1}{c}\Delta\rho_i/\Delta(\ln T)$ ($10^{-7}\Omega$ m/at.%)	$\Delta\rho_{\min}$ ($10^{-9}\Omega$ cm)	$T_{\min}$ (°K)	$-J_{sf}$(de Gennes) (eV)	$-J_{sf}$(CEF) (eV)
*Au*Yb	*A*	0.22	0.69	0.23	5	0.49	0.47
	B	1.00	3.13	1.30	6.5	0.82	0.74
	C	2.46	7.69	2.81	6.5	1.09	1.00
	D	4.94	15.44	7.42	8.5	1.37	1.26
*Ag*Yb	*A*	0.30		0.39	6.5		
	B	0.13	0.46	0.20	6.5		
	C	0.02	0.07	0.01	4.5		
	D	0.28	1.00	0.31	5.5		
	E	0.17	0.61	0.16	5		

density of states at the Fermi level, deduced from specific-heat measurements[9] is $\eta(\epsilon_F)=0.15$ states spin^{-1} eV^{-1} atom^{-1} and for the Yb^{3+} ion $g=\frac{8}{7}$ and $j=\frac{7}{2}$.

The wide dispersion of the J_{sf} values due to the thermal treatments makes it difficult to compare our results with those obtained by other authors. The only value of J_{sf} deduced previously from resistivity measurements is that of Murani[8] who finds $J_{sf}=-0.14$ eV; however, when we calculate J_{sf} from his experimental results, and using the same formulas given by him, the result is $J_{sf}=-0.63$ eV. We should mention that in both cases the interaction Hamiltonian is taken without a factor 2 ($H=-J_{sf}\vec{s}\cdot\vec{S}$).

Previously, two values of J_{sf} has been obtained by EPR in *Au*Yb. The first, -0.22 eV, was obtained by Williams *et al.*[10] and the second, -0.85 ± 0.2 eV, by Tao *et al.*[6] Measurements using the Mössbauer effect[7] give $J_{sf}=-0.55$ eV. All of these values were obtained by taking into account the crystalline electric field (CEF) splitting, leading to the ground Γ_7 doublet. In the case of resistivity measurements, the CEF effect can be taken into account by replacing in the Kondo formula the $(g-1)$ factor by $g_{\mathrm{eff}}(g-1)/g$ and j by $S_{\mathrm{eff}}=\frac{1}{2}$, where $g_{\mathrm{eff}}=3.428$ for Yb^{+3}. Doing this we found a new set of values for J_{sf} which does not differ significantly from those calculated by using the de Gennes factor simply. The two sets of J_{sf} (de Gennes) and J_{sf} (CEF) for *Au*Yb can be compared in Table I. We believe that the spread of the results may be due to the metallurgical state of the alloys used by the authors mentioned above. Nevertheless, the comparison of our values with those obtained from local methods (like EPR and Mössbauer experiments) must be done with great care because of the vector dependence of $J_{sf}(\vec{q})$.

The case of *Ag*Yb is puzzling. Our results indicate that ytterbium produces a resistivity minimum. There is, however, a certain uncertainty about the purity of the silver used for samples *B*, *C*, *D*, and *E* (Fig. 2). The small minima shown in Fig. 2 (curve 1) could conceivably be caused by 3*d* impurities. By analyzing the pure silver as well as a sample containing ytterbium, we have checked for traces of Mn or Fe, which are known to cause such minima.[11,12] In both cases we obtain 2 ppm of Mn and 47 ppm of Fe. We should mention that the 5-N silver which we used in these alloys, although it came from the same supplier, was an earlier batch than that used to make the alloys referred to later in this paper. To show the comparison, we took the sample represented on Fig. 4 ($x=0$), remeasured its resistivity, and plotted the results on Fig. 2 (sample *A*). Since the unwanted impurity (namely Fe) has presumably the same concentration in both the pure silver and the alloy samples, it should be possible to subtract the two sets of data to obtain the resistivity $\rho_i(T)$, characteristic of ytterbium. When this is done, the results (Fig. 2, curve 3) show that the minima persists.

Contrary to the case of *Au*Yb, the heat treatment of *Ag*Yb does not seem to be of great importance, except that the homogenization before rolling reduces the resistivity minimum almost to zero.

The existence of a Kondo effect in *Ag*Yb is at variance with previous experimental results and theoretical predictions, except that very recently Murani[13] measured the resistivity of *Ag*Yb and found a minimum which he attributes to a Kondo effect. We believe that it is premature to conclude, on the basis solely of these two pieces of work, that a Kondo effect undoubtedly exists in this system. Further experiments should help to clear up this point, and they are presently under way in this laboratory.

B. Yttberium dissolved in $\mathrm{Au}_x\mathrm{Ag}_{100-x}$

After their fabrication, no thermal treatment

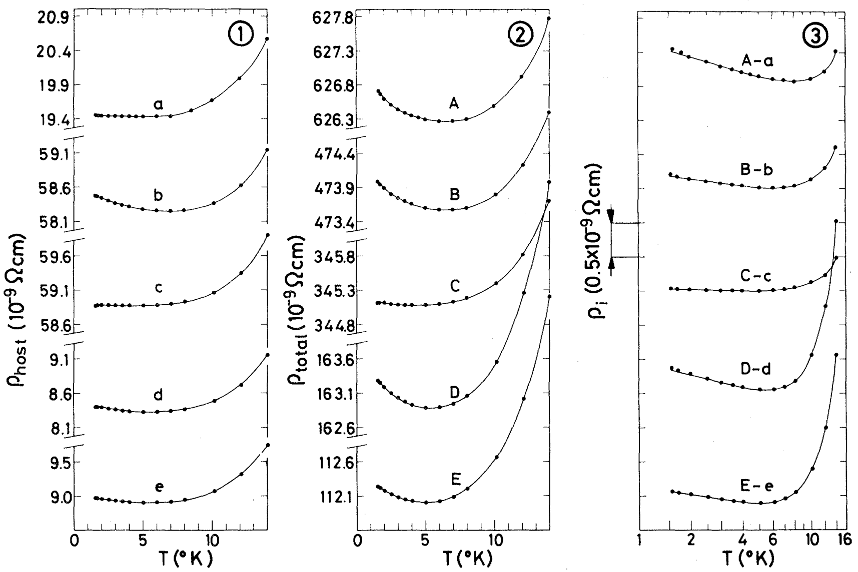

FIG. 2. As in Fig. 1 but for $Ag_{99.5}Yb_{0.5}$ (nominal concentration), except that here the heat treatments are (A, a) as-cast (these are the same samples described in Fig. 4, $x = 0$, which were remeasured in this series); (B, b) as-cast; (C, c) 1 week at 560 °C before coled working; (D, d) 1 h at 560 °C after coled working; and (E, e) 1 week at 560 °C before coled working followed by 1 h at 560 °C afterwards. All samples containing Yb (except A) were prepared with triply distilled high-purity Yb, while sample A was prepared with Yb or 99.9% purity.

was applied to any of the alloys in this series. The relative composition of the alloys was controlled by the variation of the residual resistivity (Fig. 3). The slight asymmetry between the two curves in Fig. 3 possibly indicates that the gold-rich side contains more ytterbium in solution than the silver-rich one. This is quite possibly due to the lower solubility of ytterbium in silver. Figure 4 shows representative values of the alloys and the host material as a function of temperature, while Fig. 5 shows the logarithmic behavior of their difference $[\rho_i(\ln T)]$. In order to give a general view, the results for all the measured samples are shown in Fig. 6. These include the minimum depth $\Delta\rho_{min}$ and temperature T_{min}, and the logarithmic slope $\Delta\rho_i/\Delta(\ln T)$ as a function of the host's concentration.

It is obvious that these three curves can be used interchangeably to give a qualitative picture of the Kondo effect in this system. It is also clear, in agreement with the susceptibility results of Allali *et al.*,[5] that the Kondo effect is more pronounced on the silver-rich side, with a critical concentration of about 7.5-at.% gold. The diminution of the resistivity minimum tapers off gradually on the gold-rich side, having its most pronounced falloff between 40- and 60-at.% gold.

Up to this point, the results seem to agree with the expected behavior of ytterbium in the system. A change in the valence of Yb in going from the $^1S_0$ state in silver to the $^2F_{7/2}$ state in gold is produced gradually by the addition of gold to the silver matrix. The corresponding shift of the Fermi level splits the spins of the $4f$ level at about 7.5-at.% Au and reaches its maximum effect at about 30-at.% Au. Subsequently it decreases to its value for pure gold. This interpretation is questionable, however, because if we consider the ratio of $\Delta\rho_{min}$ for the alloys $(Au_{22.5}Ag_{77.5})_{99.5}Yb_{0.5}$ and $Au_{99.5}Yb_{0.5}$, we find a value greater than 80. Similarly, the ratio of the logarithmic slopes for the same alloys gives a value greater than 50. These enormous values of the ratio are hardly reconcilable with the simple interpretation given above; we shall see later that another mechanism contributes to the resistance minimum effect in this case.

C. Effect of annealing

Considering the weak solubility of ytterbium in these alloys, we felt it was necessary to examine the possibility of a Jaccarino-Walker-type effect.[14] One expects annealing to give an atomic rearrange-

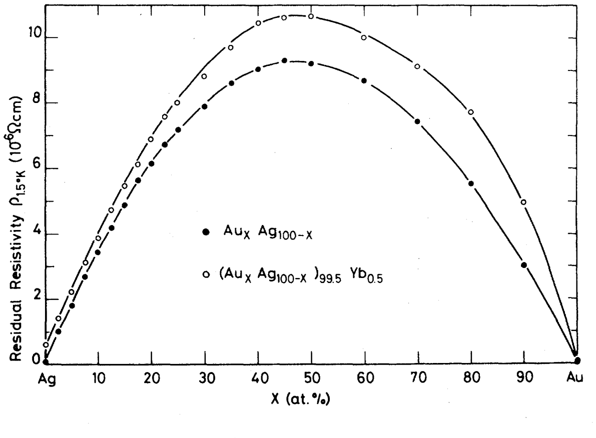

FIG. 3. Concentration dependence of the residual resistivity of the (AuAg) Yb alloys and their corresponding (AuAg) hosts.

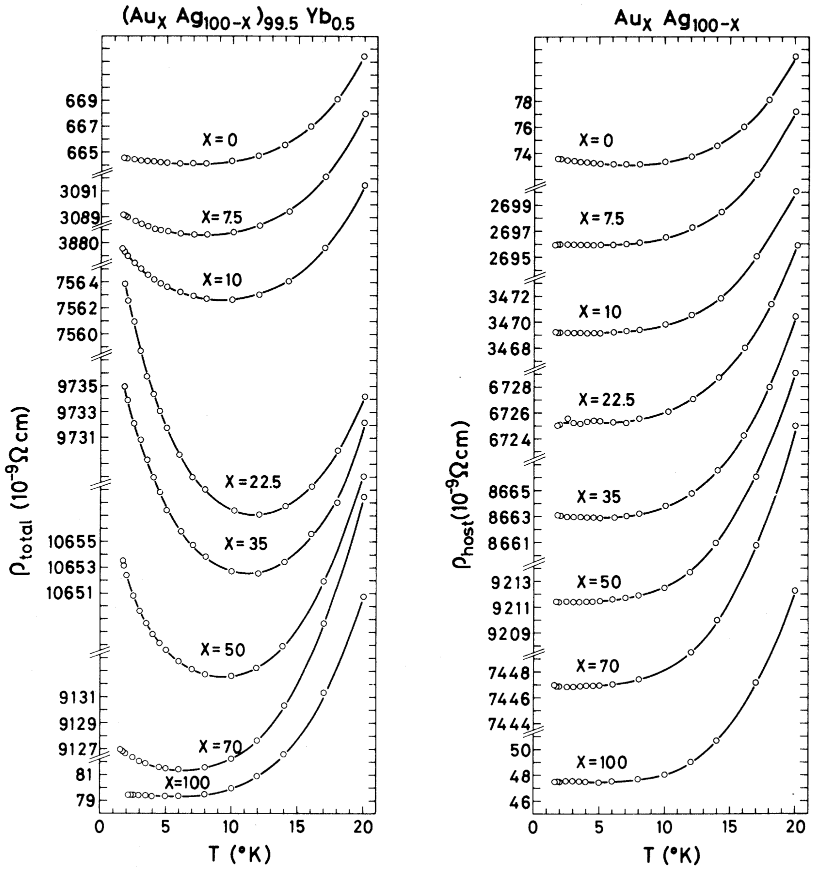

FIG. 4. Temperature dependence of the resistivities of some representative (AuAg) Yb alloys and their corresponding AuAg hosts.

ment which brings many more gold atoms close to the ytterbium atoms than the actual composition of the matrix would indicate. The bell-shaped curves on Fig. 6 would then be displaced towards the silver-rich end.

Two series of five samples in the concentration range which includes the bell-shaped part of Fig. 6 were annealed separately at 560 and 850°C and then measured. Figure 7 shows the total resistivity ρ_{tot} versus temperature, and the impurity resistivity ρ_i versus the logarithm of the temperature. It is clear that the annealing changes the properties of the system considerably. The most remarkable fact is the marked drop in the resistivity at about 3°K for the samples annealed at 560°C. Qualitatively this effect shows a maximum for 30-at.% gold, which coincides with the region of the maximum in the Kondo effect and disappears completely at about 60-at.% gold.

The samples annealed at 850°C do not show this behavior, but the resistivity minimum is nevertheless considerably diminished. However, it is still most pronounced for the $x = 30$ sample.

It seems that these results cannot be explained in terms of the previously accepted behavior of

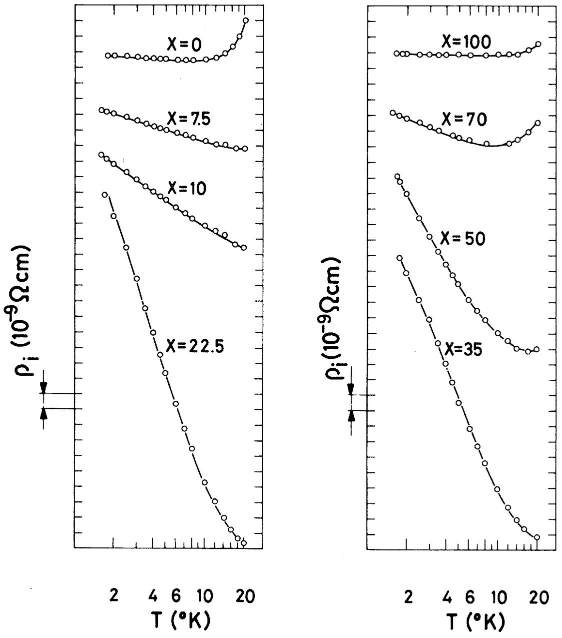

FIG. 5. Logarithmic temperature dependence of the impurity resistivity ρ_i, found for the alloys of Fig. 4.

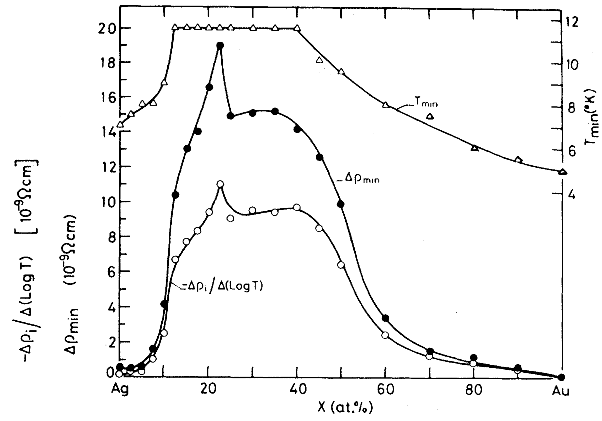

FIG. 6. Variation with the host's composition of three parameters deduced from the resistivity measurements, obtained from all the samples studied. The explanation of these parameters is given in the text.

Yb in the gold-silver system.

Measurements of the susceptibility and magnetoresistivity of the sample $x = 30$ annealed at 560 °C suggest that the sudden fall of the resistivity can be attributed to an antiferromagnetic ordering. One has to assume that this annealing favors the appearance of a distinct antiferromagnetic phase with a Néel temperature of 3.2 °K. Moreover, the similarity between the dependences on x of the Kondo effect and the magnetic ordering suggests that at least part of the resistivity minimum arises from this antiferromagnetic phase.

Annealing at 850 °C does not produce any magnetic ordering anomaly of this kind. It is expected that at such a high temperature the alloy will be single phase and stay so after quenching; note that this supression of the magnetic-phase attenuates considerably the resistivity minimum. If we now consider the $\Delta\rho_{min}$ ratio of the sample $x = 30$ before (Fig. 6) and after (Table II) annealing at 850 °C, the result is a factor close to 7, while the ratio of the logarithmic slopes gives a factor of 15. This supports the suggestion that a part of the resistance minimum effect can be attributed to the antiferromagnetic phase, which accounts in large part for the observed behavior of the system.

The comparable order of magnitude of the Kondo effect in the unannealed samples and in those annealed at 560 °C leads us to believe that the antiferromagnetic phase may be present in the unannealed state, although to a less-pronounced extent.

D. Ytterbium-rich alloys and intermetallic compounds

If the existence of the resistance minimum is to be attributed, at least in large part, to a specific phase containing Yb, one would expect that the addition of Yb would increase the size of the effect by increasing the amount of the Yb phase present. In order to test this idea we chose the alloy $x = 30$ situated in the region of the maximum of Fig. 6. The ytterbium content was varied from 0.05 to 2.0 at.%. Figure 8 shows the results obtained. As expected, the depth of the resistance minimum increases in proportion with the quantity of ytterbium present. The antiferromagnetic ordering produced by annealing at 560 °C always appears at the same temperature and is first visible even at $y = 0.2$.

The unannealed samples seem to show a slight change in slope at the same Néel temperature. Perhaps the same second phase exists in the unannealed state too, as suggested above, but badly crystallized.

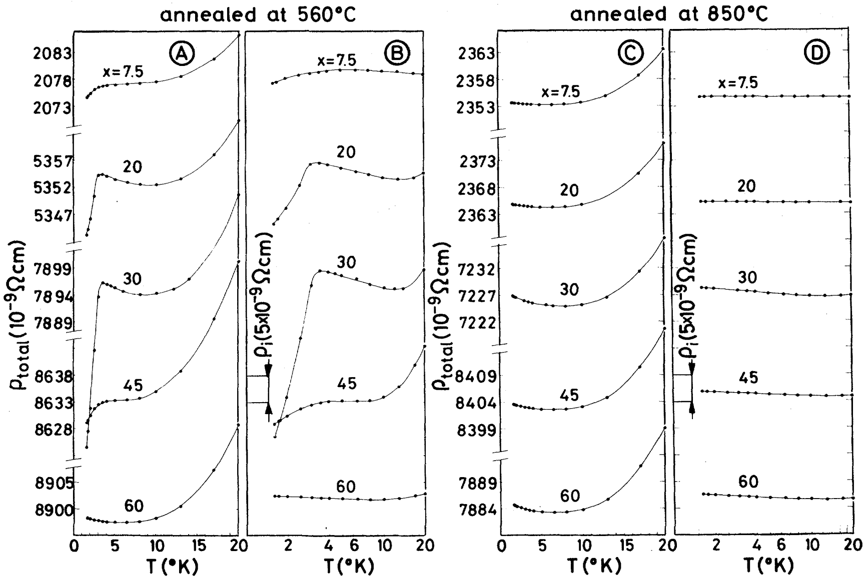

FIG. 7. Temperature dependence of the total resistivity $\rho_{tot}(T)$, and Yb contribution $\rho_i(\ln T)$, of $(\text{Au}_x\text{Ag}_{100-x})_{99.5}\text{Yb}_{0.5}$ (nominal concentration) obtained after annealing the samples at two temperatures, for 1 h, after coled working: (*A* and *B*) 560 °C; (*C* and *D*) 850 °C.

TABLE II. Parameters deduced from the measurements shown on Fig. 7 for $(Au_xAg_{100-x})_{99.5}Yb_{0.5}$ samples, which were annealed separately at 560 °C and 850 °C. The values of $\Delta\rho_{min}$ were obtained from the ρ_{total} curves, extrapolated to 1.5 °K, without taking into account the drop of the resistivity at about 3 °K, for the samples annealed at 560 °C.

Annealing temp. (°C)	alloy	$-\Delta\rho_i/\Delta(\ln T)$ ($10^{-9}\,\Omega$ cm)	$\Delta\rho_{min}$ ($10^{-9}\,\Omega$ cm)	T_{min} (°K)
560	$x=7.5$	0.65		
	20	2.61	3.4	9
	30	3.17	3.5	9.5
	45	0.20		
	60	0.48	1.0	6
850	$x=7.5$	0.15	0.5	6
	20	0.17	0.7	6
	30	1.0	2.1	7.5
	45	0.55	1.2	6.5
	60	0.62	1.5	6.5

We then attempted to identify this phase. Gold and silver are miscible in all proportions, but ytterbium, on the other hand forms numerous intermetallic compounds with Au[15] or Ag.[16] In each of its compounds with silver the ytterbium is divalent and therefore does not possess a magnetic moment. In three of its compounds with gold (namely, $YbAu_2$, $YbAu_3$, and $YbAu_4$) the ytterbium is trivalent with a magnetic moment close to the free-ion value ($\sim 4.3\mu_B$). X-ray-powder pictures taken of the sample $(Au_{30}Ag_{70})_{98}Yb_2$ before and after annealing at 560 °C show in both cases the presence of $YbAu_3$, but the blurred and indistinct lines of these pictures for the unannealed sample confirm the bad crystallization in the compound. It is quite possible that the two other trivalent compounds are also present but in less significant amounts. The method using powder pictures cannot detect amounts less than about 3%.

The same samples have been examined with the electron microprobe. The results are in agreement with those obtained from the x-ray pictures, and moreover provide some further details about the structure and the composition of the sample. Figure 9 is the surface topography found by the microprobe for the annealed sample (exactly the same thing is found for the unannealed one). A precipitated phase, shown by the clear points in the figure, appears densely distributed on the background of the matrix. The diameter of these points is approximately 5000 Å. Using the microprobe, we tried to evaluate the different concentrations of the phases present; the result was $Au_{26.8}Ag_{73.2}$ for the matrix, and $(Au_{37.6}Ag_{62.4})_{70}(YbAu_3)_{30}$ for the precipitated phase. Even though these values cannot be taken as absolute, they do suggest some interesting points. First, there is a general reduction in the gold content of the matrix when ytterbium is added. A local enrichment in gold is then produced due to its tendency to diffuse preferentially around the ytterbium. The $YbAu_3$ compound is formed inside this local environment, being richer in gold than the rest of the alloy. Probably a very small quantity of ytterbium remains as solid solution in the matrix. This could not be detected by the methods we have used.

Once the existence of the intermetallic phase was established, we tried to isolate it and to measure its resistivity in order to find its eventual contribution to the resistance minimum of the system. We chose to measure $YbAu_3$ and YbAu. The latter should not exhibit any anomaly because the

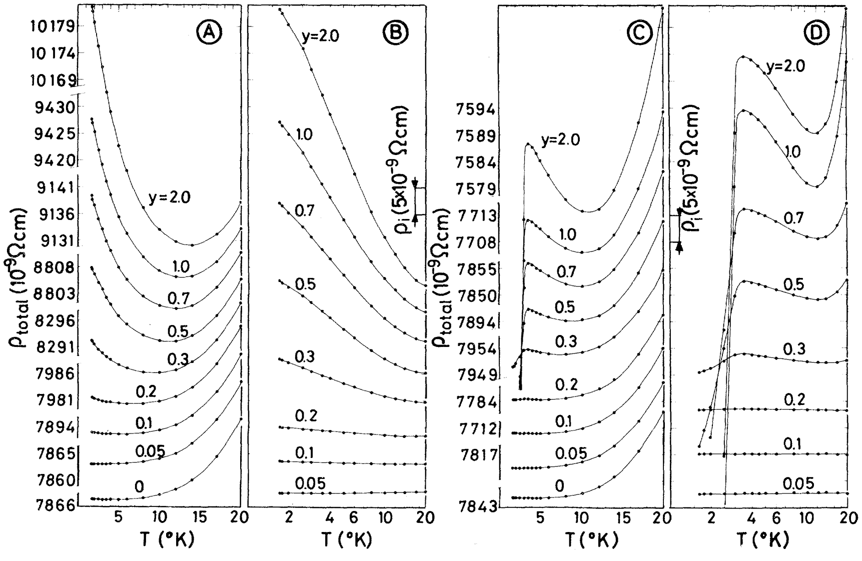

FIG. 8. As in Fig. 7 but for samples $(Au_{30}Ag_{70})_{100-y}Yb_y$: (*A* and *B*) are as-cast; (*C* and *D*) annealed for 1 h at 560 °C, after coled working.

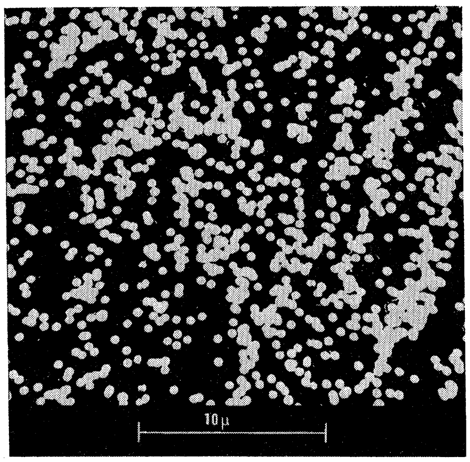

FIG. 9. Surface topography obtained by microprobe analysis of the $(Au_{30}Ag_{70})_{98}Yb_2$ sample after homogenization for 1 week at 560 °C. The clear points represent the zones containing the intermetallic compound $YbAu_3$.

Yb in it is divalent.

The large number of ordered solutions in the Au-Yb system makes it quite difficult to produce a perfectly stoichiometric sample with an arc furnace. A small evaporation of one of the components, which often occurs in an arc furnace, puts the sample into a double-phase region. Actually, the $YbAu_3$ sample we used contains an almost equal amount of $YbAu_4$, while YbAu contains likewise an almost equal amount of $YbAu_2$. Never-the less, samples prepared in this way are adequate for a preliminary measurements. Figure 10 shows the results obtained. The two samples each showed a deep resistivity minimum greater than any of those observed previously. The fact that each sample contains at least one trivalent phase explains the common features in the behavior of their resistivities. The Néel temperatures which are obtained from these results are 7 °K for $YbAu_2$ and 4.7 °K for $YbAu_4$; the corresponding value for $YbAu_3$ is masked in these data. From our previous results (see, for example, Fig. 8) one obtains 3.2 °K.

IV. CONCLUSION

Previously it has been assumed by all authors dealing with the system (AuAg)Yb that the total amount of ytterbium goes into the matrix as a solid solution. One may therefore think that the variation of the valancy of ytterbium, due to the shift of the Fermi level, is the sole cause of the observed resistance minima. Unless the correct heat treatment is applied, we have shown that a combination of the above mechanism (the shift of the Fermi level), together with that resulting from the presence of the intermetallic phase, cannot be avoided. Of these two mechanisms, the one due to the intermetallic phase dominates.

The very recent results of Nagel *et al.*[17] appeared while the present paper was being prepared. They report measurements of the EPR and electrical-resistance effects made upon samples of the same system as we have studied, but their work does not include any investigation of different thermal treatments. Their resistivity results are in qualitative agreement with ours shown in Fig. 6. In their interpretation of their EPR spectra, Nagel *et al.* have only considered the mechanism leading to the exchange integral J_{sf} for dilute solid solutions. But in view of the similarity of their resistivity results and ours, it would seem that perhaps the interpretation of the EPR results should also consider the influence of the intermetallic compounds which we have found to be present in the system.

The case of the intermetallic compounds $YbAu_x$, where Yb is trivalent ($x=2,3,4$), is itself interest-

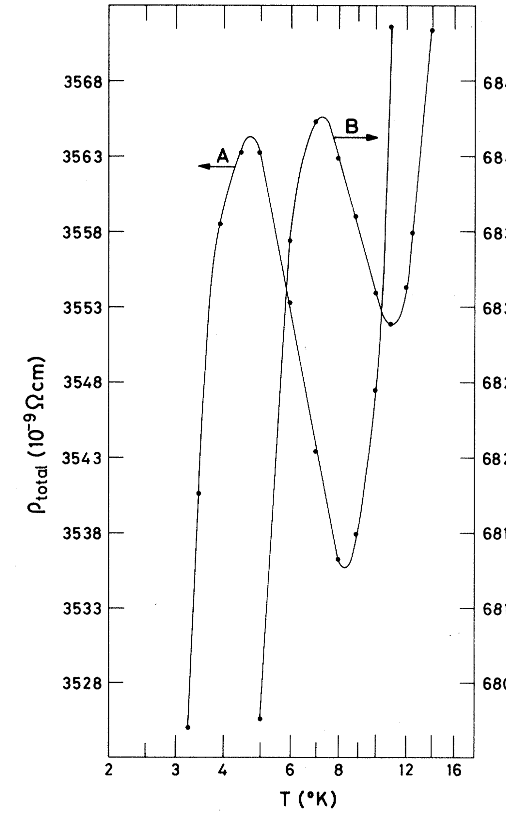

FIG. 10. Logarithmic temperature dependence of the resistivity of the Yb-Au intermetallic compounds. (a) $YbAu_3$ (+$YbAu_4$); (b) YbAu (+$YbAu_2$).

ing. The resistivity minima in this case may originate from either an intrinsic Kondo effect, as in the case of $CeAl_x$, or from critical fluctuations as the antiferromagnetic ordering transition is approached. For an intrinsic Kondo effect, the influence of the crystalline field leading to the Kondo sidebands has to be considered (as was done by Maranzana[18–20] for the $CeAl_x$ compounds). To decide between these two possibilities, we are continuing our investigations of the $YbAu_x$ intermetallic compounds using perfectly stoichiometric single-phase samples; particular emphasis is being placed upon neutron-diffraction studies.

ACKNOWLEDGMENTS

The authors would especially like to extend their thanks to Professor R. Orbach and Professor C. M. Hurd, the first for his ideas and suggestions concerning the interpretations of their results on the intermetallic compounds, and the second for his valuable help in the final phase of the preparation of this paper. They would also like to thank Professor S. Steinemann and his group at the Université de Lausanne for his measurement of our samples with the electronic microprobe. Finally, they thank M. R. Cartoni for his technical assistance during this work.

---

[1] P. E. Rider, K. A. Gschneidner, and O. D. McMasters, Trans. AIME 233, 1488 (1965).

[2] J. Bijvoet, A. J. Van Dam, and Flora Van Beek, Solid State Commun. 4, 455 (1966).

[3] D. Gainon, P. Donzé, and J. Sierro, Solid State Commun. 4, 151 (1967).

[4] J. Boes, A. J. Van Dam and J. Bijvoet, Phys. Lett. A 28, 101 (1968).

[5] V. Allali, P. Donzé, D. Gainon, and J. Sierro, J. Appl. Phys. 41, 1154 (1970).

[6] L. J. Tao, D. Davidov, R. Orbach, and E. P. Chock, Phys. Rev. B 4, 5 (1971).

[7] F. Gonzalez Jimenez and P. Imbert, Solid State Commun. 11, 861 (1972). The value of J_{sf} given in this paper was corrected later by the same authors in Solid State Commun. 13, 85 (1973).

[8] A. P. Murani, Solid State Commun. 12, 295 (1973).

[9] W. S. Corak, M. P. Garfunkel, C. B. Satterthwaite, and A. Wexler, Phys. Rev. 98, 1699 (1955).

[10] The value $|J_{sf}| = 0.03$ eV given by L. L. Hirst, G. Williams, D. Griffiths, and B. R. Coles [J. Appl. Phys. 39, 844 (1968)] was corrected later by the two first authors to $|J_{sf}| = 0.22$ eV [Phys. Rev. 185, 407 (1969)].

[11] D. Jha and M. H. Jericho, Phys. Rev. B 3, 147 (1971).

[12] T. A. Kitchens, W. A. Steyert, and R. D. Taylor, Phys. Rev. 138, A 467 (1965).

[13] A. P. Murani, Solid State Commun. 14, 199 (1974).

[14] V. Jaccarino and L. R. Walker, Phys. Rev. Lett. 15, 258 (1965).

[15] A. Iandelli and A. Palenzona, J. Less-Common Metals 18, 221 (1969).

[16] A. Palenzona, J. Less-Common Metals 21, 443 (1970).

[17] J. Nagel, S. Hüfner, and M. Grünig, Solid State Commun. 13, 1279 (1973).

[18] F. E. Maranzana, Phys. Rev. Lett. 25, 239 (1970).

[19] H. T. Van Daal, F. E. Maranzana, and K. H. J. Buschow, J. Phys. 32, 424 (1971).

[20] F. E. Maranzana and P. Bianchessi, Phys. Status Solidi 43, 601 (1971).

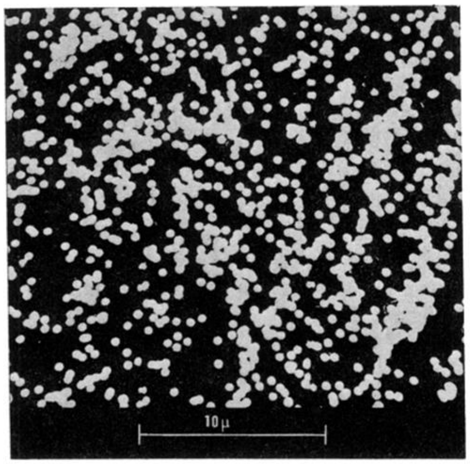

FIG. 9. Surface topography obtained by microprobe analysis of the $(Au_{30}Ag_{70})_{98}Yb_2$ sample after homogenization for 1 week at 560 °C. The clear points represent the zones containing the intermetallic compound $YbAu_3$.