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NUCLEAR-INDUCED RESISTIVITY DROP IN THE KONDO ALLOY $\text{Au-}^{171}\text{Yb}$

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Résumé. — La résistivité électrique d'alliages dilués monoisotopiques d' $\text{Au-}^{171}\text{Yb}$ et de $\text{Au-}^{174}\text{Yb}$ a été mesurée jusqu'à 18 mK. Dans l' $\text{Au-}^{171}\text{Yb}$, on observe une chute de résistivité en dessous de 90 mK. On montre que cet effet résulte du couplage hyperfin magnétique dans l' ^{171}Yb qui lève la dégénérescence des niveaux électronucléaires en produisant un état fondamental singulet et un état excité triplet. On propose un modèle simple qui rend compte des résultats expérimentaux.

Abstract. — The electrical resistivities of monoisotopic dilute alloys of $\text{Au-}^{171}\text{Yb}$ and $\text{Au-}^{174}\text{Yb}$ have been measured down to 18 mK. A resistivity drop is observed in $\text{Au-}^{171}\text{Yb}$, below 90 mK. This effect is shown to result from the magnetic hyperfine coupling in ^{171}Yb which splits the electro-nuclear levels into a singlet ground state and a triplet excited state. A simple model is proposed which accounts for the experimental results.

1. Introduction. — In dilute alloys, the Kondo effect is very sensitive to any kind of interaction experienced by the local spin S ; in the case of 4f impurities, such an interaction can result for instance from the spin-orbit coupling or from the crystal field. A similar phenomenon is also observed when different impurities are coupled via RKKY interactions. All these situations have already been investigated both experimentally and theoretically. However little work has been devoted up to now to the effect of the coupling between the electronic spin and the nuclear spin on the properties of a Kondo impurity. To allow a study of this phenomenon, a system should exhibit both hyperfine and Kondo properties in the same temperature range. This condition is met in the mono-isotopic dilute alloy $\text{Au-}^{171}\text{Yb}$.

A previous study [1] of specimens prepared with isotopes of ytterbium showed that in $\text{Au-}^{171}\text{Yb}$ (nuclear spin $I(171) = 1/2$) the magnetic hyperfine coupling causes very unusual phenomena to occur in the 10^{-1} K range: below 50 mK, the effective spin S_{eff} ($S_{\text{eff}} = 1/2$) defined inside the crystal field ground state Γ_7 and the nuclear spin I experience antiparallel coupling, and the ground state of the total electro-nuclear system in zero field is a non-magnetic singlet $F=0$ ($F = I + S$). This complete compensation of the electronic spin by the nuclear spin results in a Van Vleck-like temperature-independent susceptibility below 50 mK and an *electro-nuclear* Schottky anomaly in the specific heat, associated with the thermal population of the excited triplet.

In addition to these ionic properties, Au-Yb alloys exhibit some other features which are typical of a Kondo system: both the impurity relaxation rate deduced from Mössbauer experiments [2] and the electrical resistivity [3, 4] give logarithmic anomalies below 10 K. However, the $\log T$ term in the resistivity is very weak and the variation between 1.5 and 4.2 K is roughly three orders of magnitude smaller than the residual resistivity at 4.2 K. All these results are consistent with the assumption of a very low value of T_K ($T_K < 1$ mK): no deviation from an ionic behaviour is observed in the static properties (magnetization, nuclear orientation [5], specific heat) which are sensitive to the Kondo effect only in the range of T_K or below, whereas dynamic properties like the resistivity and the relaxation rate exhibit anomalies at temperatures several orders of magnitude larger.

To summarize, the hyperfine coupling certainly occurs in $\text{Au-}^{171}\text{Yb}$ at a temperature $T_{\text{hf}} \gg T_K$ and we can expect the resistivity to deviate from a simple Kondo variation around T_{hf} . On the contrary, in the alloy $\text{Au-}^{174}\text{Yb}$, the electronic properties can be observed without hyperfine coupling since the nuclear spin of ^{174}Yb is $I(174) = 0$. The existence of two stable isotopes of ytterbium with nuclear spins $I = 1/2$ and $I = 0$ respectively will prove very convenient for analysing hyperfine effects in Au-Yb alloys.

2. Experiments. — In this work, we have studied two monoisotopic samples $\text{Au-}^{171}\text{Yb}$ and $\text{Au-}^{174}\text{Yb}$, both of them containing about 400 at. ppm of ytter-

bium. The choice of this concentration is justified by the necessity of obtaining an appreciable thermal variation of the resistivity while keeping the alloy dilute enough regarding RKKY interactions.

In order to prevent any contamination, our specimens were melted in a high vacuum, partial levitation furnace, then cold worked into wires of 0.28 mm diameter and about 25 cm length, each operation being followed by etching. A serious metallurgical difficulty already encountered by several authors [4] is the Kondo effect of iron impurities, which can never be entirely eliminated. It is known that one atomic ppm of iron dissolved in gold gives approximately the same logarithmic slope as 350 at. ppm of ytterbium [4, 6] which implies that great care must be taken to introduce as little iron as possible during the preparation of the wires. However, the impurities added by rolling cannot be considered dissolved since the samples were never annealed. Thus, the concentration of iron given by the activation analysis (see Table I) should be considered as an upper limit of the amount of dissolved impurities which actually scatter the conduction electrons. We shall refer to this argument in the discussion of our experimental results.

The concentration of ytterbium was deduced from the saturation magnetization up to 100 kOe since this method has been shown previously [4] to give excellent results in this system. The value of the saturation moment required for this analysis ($\mu_{\text{sat}} = 1.69 \mu_B$) was taken from reference [4]. All the characteristics of the samples are summarized in table I.

TABLE I

Summary of some parameters concerning the $\text{Au-}^{171}\text{Yb}$ and $\text{Au-}^{174}\text{Yb}$ samples

Specimen	I	A_s (mK)	c_{Yb} at. ppm	c_{Fe} (analysed) at. ppm	c_{Fe} (calculated) at. ppm	$\rho(4.2)$ nΩcm
$\text{Au-}^{171}\text{Yb}$	1/2	+125	415	6-15	1.8	300
$\text{Au-}^{174}\text{Yb}$	0	—	395	3.5	3.9	288

The experiments were performed in the mixing chamber of a dilution refrigerator down to 18 mK in continuous operation. The resistivities were measured by a four-wire a.c. method using a commercial SQUID system. The best accuracy is obtained at the maximum frequency of the bridge ($\nu \approx 160$ Hz) but it was verified that the results are independent of ν within experimental precision. The reproducibility of the measurements over a period of five months including several thermal cycles up to room temperature was found equal to a few 10^{-4} whereas the sensitivity of the detection system is about 10^{-5} . However, the absolute value of R may display systematic errors up to 1 %.

The temperature was deduced from the susceptibility of a cylinder of powdered CMN (height = dia-

meter) measured by a mutual inductance method and calibrated against the vapour pressure of ^4He between 1.2 K and the lambda point.

3. **Results.** — In figure 1, the difference $\rho - \rho(4.2 \text{ K})$ for the two samples is plotted *vs* $\text{Log } T$ between 4.2 K and 18 mK ⁽¹⁾.

FIG. 1. — Variation of the resistivity of $\text{Au-}^{174}\text{Yb}$ and $\text{Au-}^{171}\text{Yb}$ below 4.2 K.

The most striking feature of these results is the resistivity maximum observed in $\text{Au-}^{171}\text{Yb}$ ($I = 1/2$) at 90 mK in contrast to the monotonic decrease of ρ with T in $\text{Au-}^{174}\text{Yb}$ ($I = 1/2$). This maximum cannot be ascribed to iron impurities since an ordering temperature of about 90 mK would require a concentration of iron higher than 100 at. ppm [6]. Such a high value is ruled out by the activation analysis ($c_{\text{Fe}} \leq 15$ at. ppm) and the very weak logarithmic slope observed between 0.1 K and 4.2 K ($c_{\text{Fe}} < 3$ at. ppm).

Neither can RKKY interactions between ytterbium ions account for this maximum since both samples have the same concentration of ytterbium but only $\text{Au-}^{171}\text{Yb}$ gives a maximum. Moreover, susceptibility measurements [1] indicate that the ordering temperature for 400 at. ppm is less than 7 mK.

From this discussion, we conclude that the resistivity maximum in $\text{Au-}^{171}\text{Yb}$ is an intrinsic, one impurity effect. Thus, the difference between the two samples must be related to the fact that the nuclear spin is 0 in ^{174}Yb whereas it is $1/2$ in ^{171}Yb . These effects are actually the counterpart in resistivity of the Van Vleck-like susceptibility reported previously [1].

To explain this result qualitatively, we can assume that in ^{171}Yb , since the strong antiparallel coupling of S and I below 50 mK removes the last degrees of freedom of the local spin, no channel is available in

⁽¹⁾ It should be kept in mind that the variation of ρ is much smaller than the value $\rho(4.2)$ which does not appear on the diagram (see table I).

the ground state $I = 0$ for the spin flip scattering of a conduction electron. A very similar effect was reported previously in the compound $\text{La}_{1-x}\text{Pr}_x\text{Sn}_3$ [7] but in that case, the singlet ground state was produced by the crystal field acting on the electronic angular momentum $\mathbf{J}$ and the resistivity drop occurred above 1 K.

Before trying a more quantitative interpretation of the resistivity anomaly in $\text{Au-}^{171}\text{Yb}$ we must correct our results for the contribution of iron impurities. Their concentration was estimated ⁽²⁾ from the slope between 1.5 K and 4.2 K after subtracting the term due to ytterbium

$$\left(\frac{d\rho}{d \log T} = -1.21 \text{ n}\Omega \cdot \text{cm/at \% Neper [4]} \right),$$

with the one-impurity resistivity of iron in gold taken from the reference [6], namely

$$\rho_{(\text{n}\Omega \cdot \text{cm/at ppm})} = 1.28 \left[1 - \cos \frac{2.81 \pi}{5} \times \cos \left(\frac{\pi}{2} + \arctg \frac{\ln(T/T_K)}{\pi \sqrt{S(S+1)}} \right) \right] \quad (1)$$

with $T_K = 0.392$ K and $S = 0.497$. This correction results in the curves shown in figure 2. The experi-

FIG. 2. — Variation of the resistivity of $\text{Au-}^{174}\text{Yb}$ and $\text{Au-}^{171}\text{Yb}$ below 4.2 K corrected for the contribution of iron impurities. The solid curves are the results of the calculation for three values of T_1 .

⁽²⁾ We have to use this indirect determination because we cannot rely on the analysed concentrations of iron which obviously contradict the smaller logarithmic slope observed in the $\text{Au-}^{171}\text{Yb}$ sample (See table I). A similar difficulty was already encountered in 5 N gold samples containing a few atomic ppm of iron (F. Lapierre and J. Teixeira, private communication). A possible reason for this apparent reduction of the impurity resistivity has been suggested above.

mental points below 40 mK have not been shown since the contribution of iron has not been measured below this temperature. Due to the rather crude procedure we had to use for determining the amount of iron impurities, the data shown in figure 2 may be affected by a fairly large uncertainty. However, we emphasize that the main features of the curves are qualitatively the same as in figure 1; in particular, the position of the maximum remains unchanged. Only the amplitude of the resistivity drop in $\text{Au-}^{171}\text{Yb}$ is strongly enhanced in comparison with the Kondo term.

4. Mathematical treatment. — In the framework of an s-f exchange model, the hamiltonian of the system $\text{Au-}^{171}\text{Yb}$ can be written ⁽³⁾

$$\mathcal{H} = nV - 2JS_{\text{eff}} \cdot \mathbf{s} + A_s S_{\text{eff}} \cdot \mathbf{I}. \quad (2)$$

The first two terms represent respectively potential and exchange scattering. The third one is the magnetic hyperfine interaction with $A_s = 0.125$ K in the crystal field ground state Γ_7 of ^{171}Yb [1]. This hamiltonian is very similar to that used by Matho and Beal-Monod [9] to describe the formation of pairs of impurities in Kondo systems such as Au-Mn , Ag-Mn , etc... :

$$\mathcal{H} = (n_1 + n_2) V - 2J[\mathbf{S}_1 \cdot \mathbf{s}_1 + \mathbf{S}_2 \cdot \mathbf{s}_2] - WS_1 \cdot S_2. \quad (3)$$

In this expression, $\mathbf{S}_1$ and $\mathbf{S}_2$ are two equal spins, just like $\mathbf{I}$ and $\mathbf{S}_{\text{eff}}$ in ^{171}Yb . The only difference with the present case is the fact that both $\mathbf{S}_1$ and $\mathbf{S}_2$ are coupled to the conduction electrons. However the coherent scattering of two impurities produces only negligible corrections to the final expression for $R(T)$. A straightforward transposition to the present case gives :

$$\rho = R_V + \frac{4}{3} \bar{R} \Sigma^2 \left(\frac{-A_s}{k_B T} \right) \times \log \left[\frac{T_1}{\sqrt{T^2 + (0.77 A_s/k_B)^2}} \right], \quad (4)$$

with

$$\Sigma(x) = \frac{3}{2} \left(1 + \frac{x}{e^x - 1} \right) / (e^{-x} + 3). \quad (5)$$

⁽³⁾ It has been shown by Cornut and Coqblin [8] that a correct treatment of the Kondo effect in rare earth compounds must take into account both spin and orbital exchange scattering, and include crystal field effects. In contrast with the usual JS_s model, their calculation results in the possibility of transitions with $\Delta M \neq \pm 1$ (with $M = \langle J_z \rangle$). However, since we are only interested in the Γ_7 crystal field ground state which is equivalent to a spin 1/2, we will not be concerned with this difference between the two models.

R_V is the potential scattering resistivity; the parameters $\bar{R}$ and T_1 are defined in reference [9] (eqs. (84) and (39)).

In eq. (4), R_V gives a temperature-independent contribution, $\bar{R}$ is the coefficient of the logarithmic slope in the limit $T \gg A_s/k_B$ (we take it equal to $-1.21 \text{ n}\Omega\text{cm/at } \%$ Neper, that is about

$$4.85 \times 10^{-2} \text{ n}\Omega\text{cm/Neper}$$

for 400 at. ppm of ytterbium), and T_1 accounts for the amplitude of the resistivity drop below the temperature of the maximum. The result of the calculation is shown in figure 2 for three values of T_1 . Because of the large uncertainty resulting from iron contamination, the result of such a fit should not be taken too quantitatively. However we emphasize that the position of the maximum is essentially dependent on the value of A_s which is not an adjustable parameter in our model and therefore, the very good agreement observed strongly supports the current interpretation. Moreover, the amplitude of the resistivity drop below the maximum provides a semi-quantitative estimation for the spin-flip contribution to the resistivity namely $5 \text{ n}\Omega\text{cm/at. } \%$ at 1 K. Such a small

value — only 0.07 % of the total resistivity at this temperature — is in reasonable agreement with an independent evaluation from the work of Cornut and Haen [4] at higher temperature, namely

$$\rho_{\text{spin-flip}} \sim 10 \text{ n}\Omega\text{cm/at } \% .$$

5. Conclusion. — In conclusion, this study of $\text{Au-}^{171}\text{Yb}$ provides the first experimental evidence for the influence of the nuclear spin of an impurity on the spin-flip resistivity of a dilute alloy. Even though this work was devoted to a striking case where a complete compensation of the electronic spin by the nuclear spin occurs below 0.1 K, a variety of situations can actually be found [10], depending on the sign of J , on the magnitude of the potential scattering and eventually on the relative values of T_K and T_{hf} . These results open a new field for dilute alloy resistivity studies at very low temperatures.

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