KONDO EFFECT AND RKKY COUPLING IN PdHx(α') Fe

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Résumé.- Le comportement du fer dans Pd H_x (α') a été étudié par effet Mössbauer sous champ magnétique. Pour une faible concentration en fer, l'effet Kondo et l'effet d'interaction entre impuretés ont pu être séparés. Le moment magnétique reste constant dans un grand domaine de compositions, tandis que la température Kondo croît avec la concentration en hydrogène.

Abstract. The Mössbauer effect in magnetic field is used to investigate the behaviour of Fe in Pd H_x (α'). It is shown that the single moment Kondo and impurity interaction effects can be separated. It is found that the magnetic moment remains constant for $0.53 < x < 0.7$ but that the Kondo temperature increases with hydrogen content.

The existence of Kondo effects and spin glass properties in $\underline{PdH_x}$ M alloys (M = transition metal impurity) has been known for some time /1,2,3/. In PdH_x, the role of the hydrogen is to progressively fill the 4d band of Pd, raising the Fermi energy into the 5s band and yielding an alloy with many of the electronic properties of Ag /4/. At ambient temperature PdH_x in the range of H/M = x from about 0.01 to about 0.57 /4/ exists as a two phase mixture $\alpha + \alpha'$. (In the literature, α' is frequently denoted as β). It has been found that the properties of $\underline{PdH}$ (α) Fe are very similar to those of $\underline{PdFe}$; Kondo and spin glass effects have been detected in $\underline{PdH}$ (α') Fe /5/.

The advantages of using hyperfine techniques to determine the Kondo temperature T_K are twofold. First we can obtain T_K at experimental temperatures $T_{mes} > T_K$ ($T_K \sim 1$ K). Second we can separate the single impurity Kondo from impurity interaction effects. Since only Fe has a convenient Mössbauer isotope in the first transition metal series, we are here concerned with PdH_x (α') $Fe_{0.003}$.

The technique used for preparing the $\underline{PdFe}$ alloy (supplied by I.A. Campbell have been described elsewhere /6/. The electrolytic charging with hydrogen was done at room temperature for H/M < 0.7 and at dry ice temperatures for H/M > 0.7 /7/.

Spectra were taken in a Mössbauer cryostat fitted with a 60 kOe magnet. Typical results are shown in Figure 1 a where the solid lines are the calculated fits to variable-width Lorentzians. From the single line spectra at zero field we calculate the isomer shift. In magnetic field, the single peak is split into 4 peaks, numbered 1,3,4,6 (peaks 2 and 5 are not observed : gamma ray direction parallel to magnetic field). This splitting is proportional to the total magnetic field at the nucleus H_{mes} . A random component in H_{mes} produces the broadening observed in the outer peaks for the spectra in magnetic field, the inner peaks remaining essentially unchanged. First we discuss the measured isomer shifts and compare these with previous results. Then we discuss the analysis of the average measured field H_{mes} in terms of a Kondo moment. Third we discuss calculations of impurity interactions which justify using the average H_{mes} for the single moment.

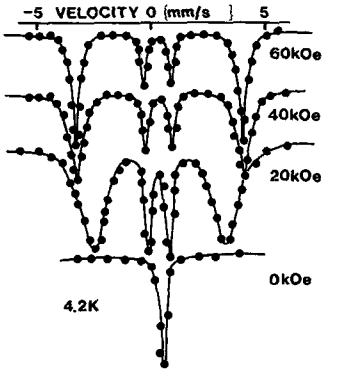

Fig. 1 : Mössbauer spectra for H/M = 0.53. For H_{ext} = 0, velocity scale expanded by a factor of 3.

Extensive hyperfine measurements were made on the compositions H/M = 0.53 and 0.7. The respective isomer shifts (IS) were 0.38 and 0.41 mm/s at 4.2 K (with respect to Fe in metallic Fe at 300 K). IS(0.53) agrees with that for α'_{min} reported in /8/ for 2 % Fe; IS(0.7) indicates α' as IS increases with H/M due to lattice expansion.

The highest H/M > 0.7 yielded two phase spectra at 77 and 4.2 K with $IS(\alpha'_{max}) = 0.62$ and $IS(\beta) = 0.89$ mm/s at 4.2 K. Some preliminary hyperfine measurements were made on a composition (IS = 0.60) close to that of α'_{max}.

H_{mes} is composed of several terms

$$\vec{H}_{mes} = \vec{H}_{ext} + \vec{H}_{HF} + \vec{H}_{VV}$$

H_{HF} is the hyperfine field produced by the local moment, $H_{HF} = H_o\, m_z\, (H_{ext})$, and is the quantity of interest. The hyperfine constant H_o is negative (due to core polarization /9/. The Van Vleck term H_{VV} is taken to be independent of temperature. It will be shown that $\beta = H_{VV}/H_{ext}$ is zero, so that $-H_{HF} = H_{sat} + H_{mes}$.

We analyse H_{HF} in terms of a modified Brillouin function :

$$H_{HF} = H_{sat}\, B_S\, (x)$$

with

$$x = \mu_{sat}\, H_{ext}/k_B(T + \theta)$$

This form has no theoretical justification but is numerically accurate for a Kondo system in the range $4T_K < T < 100\ T_K$ if $\theta = 4\ T_K$ /10/. The saturation hyperfine field H_{sat} is given by $H_o \mu_{sat}$ where $\mu_{sat} = p_{sat}\mu_B$ is the effective moment. Figures 2(a) and (b) shows H_{HF}/H_{sat} as a function of $\mu_{sat}\, H_{ext}/k_B\, (T + \theta)$ for H/M = 0.53 and 0.7 respectively. The parameters H_{sat}, $P_{sat.}$ and θ are obtained by numerical iteration (assuming $\beta = 0$) starting with the hyperfine susceptibility $\chi_{HF} = \partial H_{HF} / \partial H_{ext}$ which in the limit of zero H_{ext} is given by :

$$\chi_{HF} = \frac{\mu_B}{k_B} \frac{S + 1}{3\ S} \frac{p_{sat}\, H_{ext}}{T + \theta}$$

The extrapolation $1/\chi_{HF} = 0$ gives as shown in the inserts to figures 2 (a) and (b). From fitting H_{HF}/H_{sat} to $B_S(x)$ we obtain S = 1 and p_{sat}. Any non zero β can be detected by extrapolating χ_{HF} to $1/(T + \theta) = 0$, shown in the second inserts. The intercept $\chi_{HF} = 0$ shows that there is no temperature independent term : $\beta = 0$. Results are given in table I.

Table I

Summary of hyperfine results in Pd H_x $(\alpha')Fe_{0.003}$.

H/M	θ(K)	p_{sat}	H_{sat}(kOe)	S	H_o(kOe/μ_B)
0.53	0	4.2	- 295	1	- 70
0.7*	2	4.2	∿ - 295	1	- 70

* S=1 represents experimental results but is not the unique solution.

These results indicate that the Fe giant moment in Pd Fe (12.6 μ_B) is dramatically reduced in Pd $H_x(\alpha')$ Fe. For H/M < 0.7, μ_{sat} remains constant at about 4.2 μ_B and H_{sat} at about - 295 kOe. The sign and magnitude of H_{sat} indicate a moment of spin origin. θ increases with H/M, as was seen for T_K from resistivity measurements /1/, but not T_g of the spin glass transition which decreases /2/. Thus we conclude that θ is related to T_K: $\theta = 4\ T_K$. For H/M > 0.7, preliminary results indicate that $|H_{sat}|$ decreases, similar to what is observed at higher concentrations of Fe in the magnetically ordered phase /5, 11/. This could signal the existence of an orbital component /9/ to the moment, but further work is needed to clarify this point.

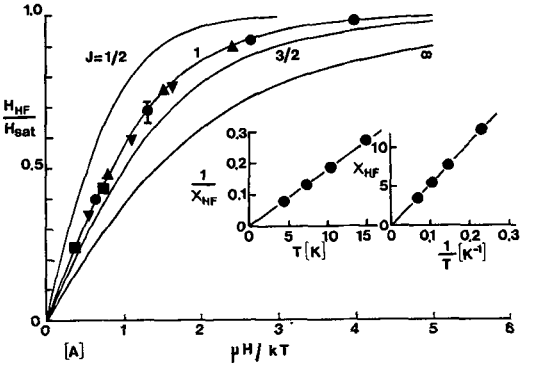

Fig. 2a.

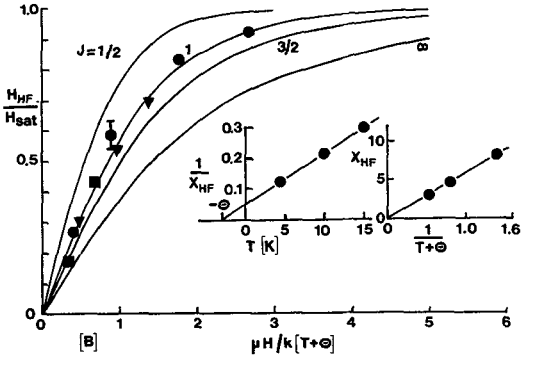

Fig. 2b.

Fig. 2 : The reduced H_{HF}/H_{sat} as a function of $\mu_{eff} H_{ext}/k_B(T + \theta)$:
● 4.2 K, ▲ 7 K, ▼ 10 K, ■15 K. B_S (x) indicated by solid lines : (a) H/M = 0.53, (b) H/M = 0.7 Inserts, see text.

Line broadening due to moment interactions has been analysed by Perez-Ramirez et al. /12/ for static RKKY coupling and by Wegener /13/ for relaxation effects. Both calculations give that the

average H_{HF} defines the behaviour of the local moment in H_{ext}, but only a relaxation time model can give such a difference in enlargement between inner and outer lines.

We interpret the line broadening as being due to the RKKY coupling between moments which leads at higher Fe concentrations to the spin glass transition. Relaxation time effects are thus similar to the relaxation effects seen in spin glasses.Further calculations will be published later.

In conclusion, it should be noted that the great advantage of hyperfine techniques applied to dilute magnetic and especially Kondo systems is the possibility to separate single moment behaviour from relaxation and interaction effects.

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