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### Anomalous magnetoresistance in 2D Pd and PdH<sub>x</sub> films

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**Résumé.** — Nous avons mesuré la magnétorésistance anormale de films minces de Pd et  $PdH_x$ , deux systèmes métalliques qui ont des structures électroniques très différentes. Dans le cas du palladium, on peut attribuer la magnétorésistance à un phénomène de localisation en présence d'un fort couplage spin-orbite; pour ce qui est du PdH<sub>x</sub> la part prépondérante est due à des fluctuations supraconductrices de type Maki-Thompson. De nos mesures, nous déduisons également les longueurs de diffusion spin-flip, électron-électron et électron-phonon et nous trouvons qu'elles dépendent fortement de la structure électronique : la longueur de diffusion spin-flip est plus grande dans le palladium, alors que les deux autres longueurs sont plus grandes dans les hydrures PdH<sub>x</sub>. Les résultats expérimentaux sont en accord raisonnable avec les théories existantes en ce qui concerne PdH<sub>x</sub>; ceci n'est pas le cas du Pd pur et nous proposons quelques modifications des théories liées à la structure électronique particulière de ce métal.

Abstract. — We report on the anomalous magnetoresistance (AMR) of thin Pd and PdH<sub>x</sub> films, two metal systems which have very different electronic structures. For pure Pd, the AMR can be attributed to a weak localization regime in the presence of a strong spin-orbit scattering rate while in PdH<sub>x</sub> it is the Maki-Thompson contribution which tends to prevail. We deduce also from these measurements the spin-flip, electron-electron and electron-phonon diffusion lengths and find them to be strongly dependent on the electronic structure : the spin-flip diffusion length is larger in Pd than in PdH<sub>x</sub>, while it is just the opposite for the two inelastic diffusion lengths. Reasonable agreement is found with theories in the case of PdH<sub>x</sub> as concerns the universal behaviour of the Maki-Thompson parameter and the absolute values of the inelastic scattering rates. For the case of Pd, refinements of the theory are needed and some suggestions are made.

#### 1. Introduction.

The transport properties of electrons in disordered metallic systems have revealed two new quantum effects. The first of these effects [1] is weak electron localization (WEL) which is due to the fact that the phase coherence of the electronic wave function is preserved over distances  $(D\tau_{\phi})^{1/2}$  (where *D* is the elastic diffusion constant and  $\tau_{\phi}$  is a characteristic life time for inelastic and spin-flip scattering) which may greatly exceed the elastic mean-free path set by the atomic disorder : the independent scattering approximation then fails with the result that positive interference in the backward direction occurs which increases the resistivity (for a review see [2]); the opposite behaviour, i.e. an increase of the conductivity, occurs [3] in the presence of strong spin-orbit scatter-

ing (weak antilocalization or WAL). The second effect results from the interplay between electronelectron interactions (EEI) and atomic disorder : the interactions are enhanced, while the Fermi level density of states and the conductivity are reduced [4, 5]; in fact there are two contributions to the EEI conductivity effect which result from the interaction between electrons with small momentum difference (diffusion channel) and large momentum difference (Cooper channel) respectively. This latter contribution becomes singular at the superconducting transition temperature  $T_{c}$  when the interactions are attractive; it then adds to the other singular contribution (Aslamazov-Larkin contribution and Maki-Thompson contribution) which originates from the superconducting Cooper pair fluctuations (for a review see [6]). Numerous experimental studies concern 2D thin films of different metals (noble metals, transition metals, semimetals) and they have confirmed the  $\ln T$  dependence predicted by the theory for both the WEL and EEI effect [2].

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An important feature allowing discrimination between the two contributions is related to the sensitivity of these effects to a magnetic field. The occurrence of an anomalous magnetoresistance (AMR) is due, in the case of WEL, to a modification of the phase of the wave function which changes the interference requirements [2]; for the EEI effects it results from a violation of the time reversal symmetry in the Cooper channel [7], while one expects no magnetoresistance from the diffusion channel as long as one can neglect the spin splitting of the conduction electrons [8]. The present work is precisely devoted to the study of this AMR for pure Pd and hydrogenated PdH<sub>x</sub> films for which the zero field behaviour [9, 10] is rather well known. The particular interest for these systems results from the possibility of modifying drastically the electronic band structure and the nature of the ground state (normal or superconducting) by increasing the concentration x without changing the geometry and the amount of disorder. It is known for instance that pure Pd (when x = 0) is a normal d metal with a high Fermi level density of states and strong repulsive electron-electron interactions resulting in a tendency towards magnetic ordering; on the other hand  $PdH_x$  with  $x \sim 0.6$ is a normal metal with an electronic structure which resembles more a noble or s-p metal with a much lower density of states at the Fermi level; superconductivity appears if one further increases the hydrogen concentration, for  $x \ge 0.7$ , and this means attractive electron-electron interactions within the Cooper channel.

Our goal is to test the sensitivity of the WEL and EEI effects to the electronic structure and to see how they interplay with a cooperative phenomenon like superconductivity; it is also possible to use these effects as a tool in order to reveal physical quantities which cannot be reached otherwise : for instance electron-electron scattering has never been observed in the resistivity of bulk PdH, samples while it will appear clearly in the AMR.

#### 2. Theory.

For 2D thin films the WEL contribution is known to take the following form [11, 12]:

$$\frac{\Delta R_{\Box}}{R_{\Box}^{2}} = \frac{R_{\Box}(H,T) - R_{\Box}(0,T)}{R_{\Box}^{2}} = \frac{e^{2}}{2\pi^{2}\hbar} \left[ -\frac{3}{2}Y\left(\frac{H}{H_{\psi}}\right) + \frac{1}{2}Y\left(\frac{H}{H_{\phi}}\right) \right]$$
(1)

 $R_{\Box}$  is the resistance per square,  $Y(x) = \ln x +$  $+\psi\left(\frac{1}{2}+\frac{1}{x}\right) \text{ is a function which varies like } x^2 \text{ for } k_{\mathrm{B}}(T-T_{\mathrm{c}}) \gg \frac{\hbar}{\tau_{\phi}} \text{ and for not too high fields i.e.}$   $x \ll 1 \text{ and like } \ln x \text{ for } x \gg 1. H_{\phi} \text{ and } H_{\psi} \text{ are critical fields which are related to the diffusion lengths } L_{\phi,\psi}, \qquad 4 eDH < k_{\mathrm{B}} T \ln \frac{T}{T_{\mathrm{c}}}.$ 

 $L_{\phi,\psi} = (D\tau_{\phi,\psi})^{1/2}$ , by the relation  $H_{\phi,\psi} = \frac{\phi_0}{4 \pi L_{\phi,\psi}^2}$ where  $\phi_0$  is the flux quantum.  $\tau_{\phi}$  and  $\tau_{\psi}$  are character ristic relaxation times given by  $\tau_{\phi}^{-1} = \tau_{in}^{-1} + 2\tau_s^{-1}$  and  $\tau_{\psi}^{-1} = \tau_{in}^{-1} + \frac{2}{3}\tau_s^{-1} + \frac{4}{3}\tau_{so}^{-1}$ ;  $\tau_{in}$ ,  $\tau_s$  and  $\tau_{so}$  are respec-tively the inelastic, spin-flip and spin-orbit relaxation times; the AMR is negative for small spin-orbit relaxation rates but changes to positive when  $\tau_{so}^{-1}$ is the largest relaxation rate. For a superconductor the EEI contribution from the Cooper channel takes the form :

$$\frac{\Delta R_{\Box}}{R_{\Box}^2} = \frac{e^2}{2 \pi^2 \hbar} \left[ \beta_{\rm MT} Y \left( \frac{H}{H_{\phi}} \right) + g_{\rm c} Z \left( \frac{H}{H_{\rm T}} \right) \right]. \quad (2)$$

The first term is the Maki-Thompson contribution due to superconducting fluctuations [13] : the parameter  $\beta_{MT}$  is directly related to the effective temperature dependent electron-electron interaction parameter  $g_c^{-1} = -\ln \frac{T}{T_c}$ . The second term is related to the modification of the density of states in relation with the attractive electron-electron interaction [7]; Z(x)is a function very similar to Y(x), but the field  $H_{\rm T}$  is given by  $H_{\rm T} = \frac{\phi_0}{2 L_{\rm T}^2}$  where  $L_{\rm T} = \left(\frac{D\hbar}{kT}\right)^{1/2}$  is the thermal diffusion length. These terms are in principle also present in a non-superconductor but they are then much weaker [14].

Thus the AMR contains four terms, but fortunately only two will be of importance in our case; previous studies [9] and the present work show that we are in the strong spin-orbit limit which means that the term  $\frac{3}{2} Y(H/H_{\mu})$  may be considered to be negligible :

formula (1) can then be replaced by 
$$\frac{\Delta R_{\Box}}{R_{\Box}^2}$$

 $- \alpha \frac{e^2}{2 \pi^2 \hbar} Y \left( \frac{H}{H_{\phi}} \right)$  where  $\alpha$  is a fitting parameter

which should be equal to -1/2. On the other hand it can be shown that the ratio  $\frac{H_{\rm T}}{H_{\phi}} = \frac{2 \pi k T \tau_{\phi}}{\hbar}$  is always much larger than one (numerical applications using the data from tables I and II give  $H_{\rm T}/H_{\phi} \gtrsim 20$ ; the second term in formula (2) can then be dropped. Finally the total AMR takes the form :

$$\frac{\Delta R_{\Box}}{R_{\Box}^2} = \frac{e^2}{2 \pi^2 \hbar} \left(\beta_{\rm MT} - \alpha\right) Y(H/H_{\phi}) . \tag{3}$$

It is this formula which is used to interpret the experimental data; it is valid not too close to  $T_c$  i.e. for  $N^{o}$  4

#### 3. Experimental.

The Pd films are deposited in a high vacuum, better than 10<sup>-8</sup> torr, by electron beam evaporation onto a glass substrate at room temperature. The thickness d is measured with a quartz crystal oscillator. We report here on a set of three films (d = 100, 50 and 25 Å) all deposited in the same run on the same substrate. Their sheet resistance  $R_{\Box} = \rho/d$  varies from 23 to 320  $\Omega/\Box$  (see Table II) and their resistivity  $\rho$ follows a Fuchs law  $\rho = \rho_0 + \frac{a}{d} (\text{with } \rho_0 \sim 15 \,\mu\Omega.\,\text{cm})$ and  $a \sim 1.900 \ \mu\Omega \text{cm}$ .Å). It is quite clear from this that our films are rather clean and that the electronic mean free path is essentially controlled by the thickness d (i.e.  $l \simeq d$ ). This demonstrates the good quality of the films as concerns their continuity and thickness homogeneity; a further check comes from the observation that the Hall coefficient  $R_{\rm H}$  is independent of d within an accuracy of  $\pm$  5 % and is very close to its value in the bulk.

These same films can be charged with hydrogen directly inside the cryostat by an electrochemical process described elsewhere [15]. The films are first charged to a maximum value  $x_m \sim 1$ ; given quantities of hydrogen are then released step by step under electrochemical control, until the pure starting Pd is recovered. The sheet resistance  $R_{\Box}$  is slightly higher in the concentrated hydrides, but they continue to follow a Fuchs law with an increased value of  $\rho_0$ but no significant variation of a; this indicates that the quality of the films is preserved after the hydrogenation procedure; further checks come from the sharpness of the superconducting  $T_{\rm c}$ , the observed finite width  $\Delta T_{\rm c} \sim 0.1$  K being most probably intrinsic. From the superconducting critical fields  $Hc_2$  we deduced the diffusion constants D (see Table I) and we checked that the product  $R_{\Box} dD$  is constant as it should (for a free electron model one has  $R_{\Box} dD = \frac{1}{3} \frac{m}{ne^2} v_{\rm F}^2$  where  $v_{\rm F}$  is the Fermi velocity and n the volume density of conduction electrons).

In order to analyse the experimental data we must be sure that the conditions of two dimensionality i.e.  $d < L_{\phi}$  is met in our films; using the data of table I, it is easy to show that this is indeed the case for the PdH, films. For the Pd films this point is more difficult to check because one does not know the diffusion constant D: but as the Fermi velocity is only two times smaller in Pd than in PdH [16], and considering that the elastic mean free path is essentially the same for both systems it appears that D is not modified by more than a factor of two and it can then be shown that the condition for two-dimensionality is still satisfied. The resistance changes were measured with a four terminal a.c. bridge in a magnetic field with H < 1 tesla produced by an electromagnet; the fractional resolution is  $10^{-6}$ ; the temperature range is 1.2 K  $\leq T \leq 20$  K.

#### 4. Results and discussion.

Before analysing our data we would like first to discuss the main features and trends we observe.

— The first point to notice is that the AMR in a perpendicular field is always positive (Figs. 1, 2, 3) at all temperatures and for all investigated concentrations x. For the pure Pd case it agrees with previous data [17, 18].

— The AMR is anisotropic, being much smaller in parallel fields; it demonstrates the orbital nature of the effect.

— The AMR is smaller in Pd than in  $PdH_x$  and in both cases it depends on the thickness (Fig. 3); this last feature has its origin in the fact that the elastic mean free path which determines the diffusion constant D is mainly set by the thickness d.



Fig. 1. — Positive magnetoresistance for 100 Å Pd film  $(R_{\Box} = 23 \Omega / \Box)$  at different temperatures. Symbols (+) are data and solid curves are the best fits using expression (3).



Fig. 2. — Positive magnetoresistance for 100 Å superconducting PdH<sub>x</sub> film. The heavy solid curve is a best fit performed at low field and using expression (3) with a field independent parameter  $\beta$ . Note the deviations at high field (see text and Fig. 6a).

Pd, 25 Å

H (Tesla)



Fig. 3. — Plots showing the influence of the film thickness on the magnetoresistance of pure Pd films and of  $PdH_x$  films with comparable critical temperatures.

0.1

— From the absence of superconductivity in pure Pd, one can conclude that the relatively small magnetoresistance stems only from WEL; the positive sign can then only be explained if one admits a strong spin-orbit scattering rate i.e.  $\tau_{so}^{-1} \ge \tau_{in}^{-1}$ : this is not an unexpected result considering the large value of the atomic number Z and if one remembers that  $\tau_{so}^{-1} \sim Z^4$ .

 $\tau_{so}^{-1} \sim Z^4$ . — The higher AMR for PdH<sub>x</sub> involves *a priori* both a WEL and an EEI contribution. Indeed we observe that the AMR increases drastically as soon as the superconducting  $T_c$  enters the experimental temperature range : the presence of Maki-Thompson fluctuations with a positive AMR is thus clearly revealed. The WEL contribution is still present and positive as shown by the results far from  $T_c$  where the Maki-Thompson contribution becomes small.

— We note also that  $\Delta R(H)$  follows quite exactly the theoretical prediction of formula (3) i.e. it varies like Y(H); this is particularly striking for the case of pure Pd (Fig. 1) where the agreement is excellent up to the highest fields used; for the superconducting hydrides (Fig. 2) progressive deviation from Y(H)begin to occur at a field which clearly depends on the ratio  $T/T_c$ : the deviations are largest and occur at lower fields as one approaches  $T_c$ . We will see below that this can be related to a field dependence of the Maki-Thompson parameter  $\beta_{\rm MT}$ . In all cases it is nevertheless possible, by a best fit procedure at low fields, to extract reliable values for  $\beta - \alpha$  and for the field  $H_{\phi}$ .

4.1 DETERMINATION OF  $\beta - \alpha$ . — For pure Pd we find that  $\beta - \alpha$  is temperature independent (Fig. 4) and its mean value is  $\beta - \alpha = 0.5 \pm 0.1$ : clearly this means that  $\beta \sim 0$  and  $\alpha = -1/2$ , i.e. we have only a WAL contribution.

On figure 4, we also plot the values of  $\beta - \alpha$  versus T for the 25 Å samples with different hydrogen contents, that is to say different  $T_c$  values. A clear divergence



Fig. 4. — Temperature dependence of the parameter  $\beta(T) - \alpha$  for 25 Å film studied at different hydrogen concentrations x and consequently different values of the superconducting critical temperature  $T_c$  (the arrows indicate the measured  $T_c$  values). The hydrogen concentrations are estimates.



Fig. 5. — Dependence of  $\beta(T/T_c) - \alpha$  as a function of the reduced temperature  $T/T_c$  for the different films with their different  $T_c$  values. Different symbols correspond to the data and the solid curve is a theoretical plot of  $\beta(T/T_c) + 0.5$ , after Larkin [13].

is now observed for the samples which have a  $T_c$ in the experimental T range. On figure 5 we plot the values of  $\beta - \alpha$  for six different samples as a function of  $T/T_c$ : all points now fall on the same curve i.e. we have a universal behaviour. The line drawn through the points is theoretical and is obtained using Larkin's relation [13] between  $g_c^{-1}(T) = -\ln \frac{T}{T_c}$  and  $\beta_{\rm MT}$ and by using  $\alpha = -1/2$ . As one can see, the agreement is excellent; it confirms measurements done on Al [19-21] but to our knowledge this is the first study where  $T_c$  is varied in a systematic manner. It appears

0.01

that the determination of  $\beta(T)$  is a very sensitive tool for detecting superconductivity with very low  $T_c(T_c < 1 \text{ K})$ : from the universal curve  $\beta(T/T_c)$ , it is possible to get  $T_c$  values which can be up to ten times smaller than the lowest experimental temperature T.

4.2 MAGNETIC FIELD DEPENDENCE OF  $\beta_{\rm MT}$ . — From figures 2 and 3 it is apparent that deviations from the ideal behaviour  $\Delta R(H) \sim Y(H)$  occur in PdH<sub>x</sub> above some kind of critical field : in fact  $\Delta R(H)$  increases less quickly with the field than theoretically expected from formula (3). The deviations depend both on the temperature and on the diffusion constant D : at a given field value they increase as T approaches  $T_c$ and they decrease as D (or the thickness) decreases. To show these trends more clearly, we define on figure 6a a critical field  $H^*$  for which  $\frac{\Delta R_{\rm th}(H^*) - \Delta R_{\rm exp}(H^*)}{AB_{\rm c}(H^*)} = 0.1$  where  $\Delta R_{\rm exp}(H^*)$  is the



Fig. 6. — a) Field dependence of the relative difference between the experimental magnetoresistance  $\Delta R_{exp}$  and the theoretical value  $\Delta R_{th}$  (solid curve in Fig. 2). The data are measured at the same temperature for two films of different thicknesses but with similar  $T_c$ . b)  $H^*(T/T_c)$  as defined in Fig. 6a for 25 Å and 100 Å thick PdH<sub>x</sub> films.

measured AMR while  $\Delta R_{\rm th}(H^*)$  is the value one obtains by extrapolating the low field Y(H) behaviour up to the value  $H^*$ ; figures 6a and 6b compare the value of  $H^*$  for two samples with different thicknesses but very similar  $T_c$  values : as can be seen  $H^*$  goes to zero as T goes to  $T_c$  and its values are larger for the thinner sample which has a smaller diffusion constant D.

These effects most probably have their origin in the field dependence of the interaction parameter  $g_c(T, H)$  which has been considered by Alt'shuler *et al.* [7] and more recently by MacLean *et al.* [22]. The observed trends as a function of T and D are qualitatively in agreement with these theories but a quantitative fit could not be obtained; the experimental deviations from the behaviour given by formula (3) are smaller than calculated theoretically, using g(T, H) given by MacLean *et al.* [22] and assuming that  $\beta$  is related to g(T, H) in the same way as it is in the low field limit.

4.3 PHASE LOSS SCATTERING PROCESSES. — On figure 7 we plot the field  $H_{\phi} = \frac{\phi_0}{4 \pi L_{\phi}^2} = \frac{\hbar}{4 eD} \frac{1}{\tau_{\phi}}$  as a function of temperature for the 50 Å sample without (x = 0)and with  $(x \sim 0.9)$  hydrogen. As is evident from the figure  $H_{\phi}$  has the form  $H_{\phi} \equiv 2 H_s + H_{in}(T) = A + f(T)$ i.e. it can be decomposed into temperature independent and temperature dependent terms. The first term A is to be attributed most probably to spin-flip scattering due to the presence of some residual magne-



Fig. 7. — Temperature dependence of the characteristic field  $H_{\phi} = 2 H_{\rm s} + H_{\rm in}(T)$  for a 50 Å film without hydrogen (pure Pd) or with hydrogen ( $x \sim 0.8$ ).

tic impurities while the second deals with inelastic scattering processes in relation with electron-electron and electron-phonon interactions.

4.3.1 The spin-flip term A. — It is interesting to note that this term is systematically smaller in Pd than in PdH<sub>x</sub> (see Fig. 7, Tables I and II) while we observe the opposite trend for the temperature dependent term  $H_{in}(T)$ . Admitting that 2  $H_s$  is due to magnetic impurities we can remark that this trend coincides with what is found in the corresponding bulk metals : all d impurities like Cr, Mn, Fe, Co have a smaller resistivity in Pd (typically ~ 2  $\mu\Omega$  cm/at % of impurity [23]) compared to PdH<sub>x</sub> (typically 5  $\mu\Omega$  cm/at % impurity [24]); in a schematic way one can say that a d impurity disturbs an s-p metal like PdH, more than it does a d metal like Pd. From the value of A, one can try to determine the approximate concentration of magnetic impurities : for the 50 Å sample of PdH, one calculates that  $\tau_s \simeq 4 \times 10^{-11}$  s from which one can estimate the resistivity  $\rho_s = \frac{m}{ne^2} \frac{1}{\tau_s}$  where *n* is the volumic density of electrons (we take  $6 \times 10^{28}$  electrons/m<sup>3</sup> corresponding to 1 electron/atom); admitting that  $\rho_s$  is proportional to the impurity concentration

we determine a concentration of  $\sim 3 \text{ ppm}$  of d impurities. This value is compatible with the starting purity of our Pd. It is interesting at this stage to note the sensitivity of the AMR to small amounts of magnetic impurities.

**4.3.2** The inelastic terms for PdH<sub>x</sub>. — We will first discuss the experiments for PdH, because it is a metal similar to some other simple metals like Cu, Ag, Al... which have already been studied elsewhere.

The analysis of the temperature dependent term is obtained by making a best fit of  $H_{\phi}(T)$  with a polynomial expansion and from this it appears that  $H_{in}(T)$ can be represented by a linear and a cubic term i.e.  $H_{\rm in} = BT + CT^3$ ; this is (precisely) the kind of dependence one expects from 2D electron-electron scattering as concerns the linear term and from electron phonon scattering for the cubic term. Table I summarizes the experimental values obtained for Band C; (in Figs. 8 and 9 we plot the temperature dependent or inelastic part of  $H_{\phi}$  and the corresponding inelastic diffusion lengths  $L_{in}^2 = \frac{\phi_0}{4 \pi H_{in}}$ 

Table I. — (PdH,
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Fig. 8. — Inelastic fields  $H_{in}$  and inelastic diffusion lengths  $L_{in}$  (right hand scale) as a function of temperature for three  $PdH_x$  films of thicknesses 25, 50 and 100 Å. Symbols (+) are the experimental points obtained from the magnetoresistance data and the solid curves are best fits using the form  $BT + CT^3$ . The values of B and C are given in table I.

We will now compare the observed values of B and Cwith theoretical expectations.

— The expression for the electron-electron life time in the 2D dirty limit is given by Abrahams et al. [25] :

$$\frac{1}{\tau_{\rm in}({\rm e-e})} = \frac{e^2}{2 \pi \hbar^2} R_{\Box} k_{\rm B} T \ln \frac{T_1}{T}$$
(4)

 $(T_1 = 1.85 \times 10^5 (k_F l)^3$ , in degrees Kelvin). For the explicit calculations we take  $l \sim d$ ,  $k_F =$  $1.22 \times 10^8$  cm<sup>-1</sup> (corresponding to 1 electron/atom). The calculated values of B are given in table I : one sees immediately that the agreement is excellent for the d = 100 Å sample, but that it is less good for the thinner samples; in fact, it appears that  $\tau_{in}^{-1}$  increases less quickly with  $R_{\Box}$  than expected from the theory,

d (Å)	$R_{\Box}$ ( $\Omega/\Box$ )	D (m²/s)	А (10 <sup>-4</sup> Т)	$\frac{B}{(10^{-4}\mathrm{T/K})}$	$C$ $(10^{-4} \mathrm{T/K^3})$	$(10^{-4} \text{ T/K})$ B (theory)	$(10^{-4} \text{ T/K}^3)$ C (theory, 3D)	$(10^{-4} \text{ T/K}^3)$ C (theory, 2D)
25	360	$ \begin{array}{r} 1.7 \times 10^{-4} \\ 3 \times 10^{-4} \\ 4.8 \times 10^{-4} \end{array} $	254	101	0.66	333	0.25	1.07
50	107		290	25.2	0.32	62.3	0.14	0.60
100	33		260	10.8	0.13	13.2	0.09	0.39



Fig. 9. — Same plot as in figure 8 for Pd films. Solid curves are again best fits using the form  $BT + CT^3$ . The values of B and C are given in table II.

a result which has also been mentioned by other authors [2], but it is interesting to note that in all cases we have B (experimental) < B (theory) which means that it is not necessary to look for a further mechanism in order to explain the absolute value of  $\tau_{in}^{-1}(e-e)$ ; one can also remark that the measured  $\tau_{in}$ (e-e) is much shorter than the corresponding time calculated in a clean 2D conductor for which  $\hbar \tau_{\rm in}^{-1}(\text{e-e}) = \frac{\pi}{8} \frac{(k_{\rm B} T)^2}{E_{\rm F}} [2]$  : at T = 10 K the calculated value would be  $10^{-8}$  s while the observed value is  $2 \times 10^{-11}$  s (for the 50 Å sample); this discrepancy emphasizes the large enhancement factor of the electron-electron interactions due to atomic disorder. — For the electron-phonon term C: we must first discuss the dimensionality of our films with respect to phonon propagation because the electronphonon interaction can lead to a cubic term either in the clean  $(l > \lambda_{\rm T})$  three-dimensional  $(d > \lambda_{\rm T})$ limit or in the dirty  $(l < \lambda_T)$  two-dimensional  $(d < \lambda_T)$ limit;  $\lambda_{T}$  is the thermal phonon wavelength; because in our case we have  $l \sim d$  it is in principle possible to switch from one case to the other by simply changing

the temperature. We calculate that 
$$\lambda_{\rm T}({\rm \AA}) \simeq 4 \left(\frac{\theta}{T}\right)$$

where  $\theta$  is the Debye temperature; crude estimates with  $\theta \sim 200$  K then show that two-dimensionality is fulfilled at 1 K for all samples but not at 10 K for the thickest one. In this reasoning we neglect the coupling of the film with its substrate which reinforces the 3D character of the phonons. For all these reasons we compare the experimental values with both the 3D and 2D theory.

The value of  $\tau_{in}(e-ph)$  in 3D (clean limit) is given by [26]:

$$\frac{1}{\tau_{\rm in}({\rm e-ph})} = 14 \ \pi \zeta(3) \left(\frac{k_{\rm B} T}{\hbar}\right)^3 \frac{\partial \alpha^2 F}{\partial \omega^2} \qquad (5)$$

 $\alpha^2 F(\omega)$  is the Eliashberg function, with  $F(\omega)$  the phonon density of states while  $\alpha^2$  is related to the electron-phonon matrix element. For a Debye model one has  $\alpha^2 F \sim \omega^2$  and one can then express  $\tau_{in}$  as a function of the electron-phonon coupling parameter

$$\lambda = 2 \int_0^{\omega_0} \frac{\alpha^2 F(\omega)}{\omega} d\omega$$

The result is :

$$\frac{1}{\tau_{in}(e-ph)} = 14 \ \pi \zeta(3) \ \lambda \omega_{D} \left(\frac{T}{\theta}\right)^{3} \tag{6}$$

 $(\omega_{\rm D}$  is the Debye frequency).

Adapting formula (6) to the 2D clean limit, we obtain :

$$\frac{1}{\tau_{\rm in}({\rm e-ph})} = 14 \ \pi^2 \ \zeta(2) \ \frac{\lambda c_{\rm s}}{d} \left(\frac{T}{\theta}\right)^2 \tag{7}$$

 $(c_{\rm s}$  is the phonon velocity).

We must multiply this formula in the dirty limit by  $q_T l$  where  $q_T = 2 \pi / \lambda_T$ . Taking  $l \sim d$  one obtains :

$$\frac{1}{\tau_{\rm in}({\rm e-ph})} = 14 \ \pi^2 \ \zeta(2) \ \lambda \omega_{\rm D} \left(\frac{T}{\theta}\right)^3. \tag{8}$$

It is interesting to note that formulae (6) and (8) are very similar and that they are independent of the thickness and the mean free path.

This is to be contrasted with  $\tau_{in}^{-1}(e-e)$  which contains the sheet resistance  $R_{\Box}$  whose value depends explicitly on *l* and *d*. Indeed we observe (Table I) that the coefficient *C* varies less rapidly with the thickness than does *B*. This seems to exclude also the possibility that  $\lambda$  itself depends on the mean free path i.e.  $\lambda \sim 1/l$ as may happen in very dirty superconductors [2].

In table I we see the rather satisfying agreement one obtains between the calculated and observed values of C if one takes  $\theta \sim 200$  K (a mean value between longitudinal and transverse phonons) and  $\lambda \simeq 0.15$  which represents the contribution of the acoustical phonons [16] to the total electron-phonon coupling parameter (the interaction with the optical phonons is neglected here as they are not thermally excited at the low temperatures considered). It is interesting at this point to compare our results to those obtained on simple metals by other authors. For instance our results are similar to those of Santhanam *et al.* [27] for Al as concerns the temperature dependence of  $\tau_{in}(T)$  as well as the kind of quantitative agreement with theories; a linear temperature dependence of  $\tau_{in}^{-1}(T)$  has also been reported for several other metals like Ag, Au, Mg, Bi ([2] and references cited therein) at low temperatures, which is compared with the theory of Abrahams *et al.* [25] for the e-e interaction; a tendency for a steeper temperature dependence is found in all cases at higher temperatures with power laws mostly around  $T^2$  or  $T^3$  but a quantitative analysis has not always been made.

4.3.3 The inelastic terms for Pd. — As is immediately visible from figures 7, 8 and 9 the temperature dependent part  $H_{in}(T)$  of  $H_{\phi}(T)$  is much larger in Pd than in PdH<sub>x</sub>, especially for the thicker samples. At first sight one might relate this to just a modification of the diffusion constant  $D \sim v_F l$ : but the Fermi velocity  $v_F$  is only two times smaller in Pd than in PdH<sub>x</sub> [16] while the mean free paths l are the same so that this cannot account for the whole difference between PdH<sub>x</sub> and Pd and more fundamental reasons must be looked for.

Just like for  $PdH_x$ , we have compared the temperature dependent part of  $H_{\phi}(T)$  with a polynomial T expansion and we obtain a reasonable fit either with a pure  $T^2$  behaviour as reported by Markiewicz et al. [18] or with a  $T + T^3$  behaviour. From the theoretical point of view a pure  $T^2$  is possible both for the e-e interaction and e-ph interaction in the clean 2D limit :  $\frac{1}{\tau_{in}(e-e)}$  is then equal to  $\frac{\pi}{8} \frac{(k_{\rm B} T)^2}{\hbar E_{\rm F}}$  which is at least 10<sup>3</sup> times too small, while  $\frac{1}{\tau_{in}(e-ph)}$  is given by formula (7) which presents a 1/d variation not observed experimentally; anyhow the clean 2D limit implies also that  $d < \lambda_{\rm T}$  and  $l > \lambda_{\rm T}$  and this conflicts with the experimental finding that  $d \sim l$  so that we reject this possibility. For these reasons, we prefer to adopt a T,  $T^3$  expansion as for PdH<sub>x</sub>. We also use it for Pd. The corresponding fits are given in figures 7, 8, 9 with the values for B and C in table II. These values of B and C are now much larger than in PdH<sub>+</sub> (Table I) and we will discuss some of the reasons which might explain this increase; for this it is necessary to consider separately the electron-electron and the electron-phonon contributions. The first reason which may increase the rate  $\tau_{in}^{-1}(e-e)$  comes from the

Table II. — (Pd).

fact that the conductivity in pure Pd is dominated by the s electrons with the possibility for them to be scattered into d states whose proper contributions to the conductivity is small by itself; this two band or s-d type of scattering means that the e-e scattering rate is possibly multiplied by a factor of the order

 $1 + \frac{N_{d}}{N_{s}}$  where  $N_{d}$  and  $N_{s}$  are respectively the d and s type Fermi density of states; another possibility stems from the existence in pure Pd of magnetic fluctuations within the d electrons (paramagnons) which may strongly scatter the s electrons. These new scattering mechanisms effects explain why a rather large e-e  $T^2$ term is observed in the resistivity of pure bulk Pd while it is not observed in  $PdH_x$  [28]; we suggest that the difference between the B values of Pd and PdH, can be explained along the same lines. For the electron-phonon term C, the difference between Pd and PdH<sub>x</sub> may result from different values of the e-ph parameter  $\lambda$ , a fact which is expected both from theoretical calculations [29] and indirectly from experiments [30] to be about three times larger in Pd; because PdH<sub>x</sub> is a superconductor while pure Pd is not. This might appear at first view to be a paradox but it should be remembered that the absence of superconductivity in Pd can precisely be attributed to the enhanced e-e interactions just discussed above while its presence in PdH, is more related to the optical H vibrations which here play no role as they are not thermally excited. Further investigations are clearly needed in order to test these ideas but it seems that it is probably not necessary to look for other types of interactions in order to understand our data both qualitatively and quantitatively. Similar measurements have been made on a few transition metals like W-Re [31] or Nb [20] : in all cases a linear T term is found which is attributed to e-e interaction,

but the e-ph term seems to be absent in W-Re and to vary like  $T^2$  in Nb; it is interesting to note that the absence of an e-ph in amorphous W-Re may indicate that a very short mean free path might suppress this term.

#### 5. Conclusions.

Our main conclusion can be stated as follows : we observe a positive AMR in 2D Pd and  $PdH_x$  films which can be interpreted as being due solely to WEL

<i>d</i> (Å)	$R_{\Box} \left( \Omega / \Box  ight)$	$2 H_{\rm s} (10^{-4} {\rm T})$	$B(10^{-4} \text{ T/K})$	$C (10^{-4} \text{ T/K}^3)$
25	322	100	184	1.63
50	90	123	90.5	1.21
100	23	72	63	0.45

in Pd in the presence of a strong spin-orbit scattering rate while Maki-Thompson fluctuations tend to prevail in PdH<sub>x</sub> as soon as the superconducting  $T_c$ approaches or penetrates the investigated temperature range. We show that this last contribution given by the term  $\beta$  behaves in a universal way if plotted as a function of  $T/T_c$ ; a saturation of the Maki-Thompson term is also observed for high enough fields which can be attributed to a field dependence of the effective Cooper channel interaction parameter. We also measure the phase loss field  $H_{\phi}$  or the corresponding diffusion length : three contributions are identified stemming respectively from spin-flip, electron-electron and electron-phonon scattering. A reasonable agreement is obtained with available theories for PdH<sub>x</sub>. As concerns Pd we observe shorter inelastic diffusion lengths compared to PdH<sub>x</sub> which cannot be explained only by a modification of the diffusion constant D; for the e-e interaction we propose that electronic band structure effects are involved in relation to s-d scattering or with diffusion on paramagnons; for the e-ph interaction we propose that the corresponding acoustic interaction is larger in Pd than in PdH<sub>x</sub>.

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