Localization, magnetism, and superconductivity in two-dimensional $Pd_{1-y}Ni_yH_x$ films

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Through localization studies we measure the spin-flip and the inelastic electron-electron scattering rates τ_S^{-1} and τ_{ee}^{-1} in $Pd_{1-y}Ni_yH_x$ two-dimensional films, a system which appears to be nearly magnetic. A strong increase of τ_{ee}^{-1}, in comparison with pure PdH, is observed for the saturated ($x\sim 1$) samples. A decrease of x leads to a further increase of τ_{ee}^{-1} as well as to a marked increase of τ_S^{-1}. We relate these effects to the growth of magnetic fluctuations on either isolated Ni atoms or on small Ni clusters. These results can also explain the decrease with Ni content of the superconducting T_c in bulk and thin films of Pd-Ni-H.

I. INTRODUCTION

One of the most interesting properties of two-dimensional (2D) metallic but disordered thin films is their anomalous magnetoresistance (AMR) associated with weak localization (WL) or weak antilocalization (WAL). It mainly allows the determination of the phase coherence breaking rate and hence the characteristic scattering rates τ_S^{-1} (spin flip), τ_{in}^{-1} (inelastic), and $\tau_{\text{s.o.}}$ (spin orbit).[1] Reviews on this problem are given in Ref. 2.

In our preceding works,[3,4] we have investigated these properties in pure Pd and PdH_x and found large differences between both systems. One of the main results concerns the inelastic electron-electron scattering rate τ_{ee}^{-1} which is found to be much larger in Pd than in PdH_x, an effect which we related to the tendency for Pd to be closer to the magnetic instability criteria $I>1$ [with $I=N(\varepsilon_F)U$, where $N(\varepsilon_F)$ is the Fermi level density of states and U the repulsive Coulomb interaction energy]. Recent theoretical works[5,6] give credit to this idea by showing that τ_{ee}^{-1} is essentially enhanced by the Stoner factor $S=1/(1-I)$ in relation to the scattering of electrons from extended magnetic fluctuations (i.e., from extended paramagnons). Other recent experimental studies[7] have shown the influence of magnetic atoms, deposited on top of localizing film, on the phase-breaking time. In the present work, we deal with 2D $Pd_{1-y}Ni_yH_x$ films, the potentially magnetic species being dissolved inside the PdH_x metal. These alloys are ferromagnetic for $x=0$ and $y>2.5$ at. % and they show well-defined localization behavior in zero magnetic field.[8,9] They become nonmagnetic and even superconducting when hydrogenated to saturation ($x\simeq 1$), indicating that *a priori* there are no long-range magnetic correlations between the Ni atoms nor localized moments on the Ni atoms. But one nevertheless expects some magnetic fluctuations, more localized as compared to the extended ones in pure Pd, which are due to a larger Coulomb energy U(Ni) on the nickel atoms. A comparable theoretical situation, with strictly localized fluctuations on isolated atoms like Ni, has been analyzed for 3D systems[10] and it was shown there that it leads to an increase of the magnetic susceptibility (i.e., $\Delta\chi\sim 1/T_f$) and of the *e-e* scattering resistivity [i.e., $\Delta\rho\sim(T/T_f)^2$], where $k_BT_f\simeq[(1-I)/IN(\varepsilon_F)]$ is the magnetic fluctuation temperature [with $I=N(\varepsilon_F)U(\text{Ni})$]. It is shown also that the dependence of $\Delta\rho$ on the local Stoner factor $S=1/(1-I)$ shows a much stronger divergence than that corresponding to the extended case (i.e., for pure Pd): It is a consequence of the fact that local paramagnons have no wave-vector dependence; this can reinforce the strength of the magnetic fluctuation effects. The situation in Pd-Ni-H_x is supposedly somewhat intermediate, presenting at the same time some extended fluctuations because U(Pd) is not zero, and local fluctuations because $U(\text{Ni})>U(\text{Pd})$. For all these reasons one expects larger τ_{ee}^{-1} rates for Pd-Ni-H as compared to PdH. The tendency towards magnetism in Pd-Ni-H_x can even be amplified by decreasing the concentration x. If x is homogeneously distributed over the sample, then one expects just an increase of the mean density of states $N(\varepsilon_F)$, which is known to be larger in Pd as compared to PdH but it cannot be ruled out that the decrease of x gives rise to slightly inhomogeneous hydrogen distributions, x being larger in the vicinity of the Pd atoms. This can be so because the atomic Ni-H interaction is more repulsive than that of the Pd-H. One expects, then, a larger increase of the Stoner factor for the Ni atoms than for the Pd atoms. Isolated Ni atoms or small Ni clusters (which exist statistically in an alloy with $y=6$ at. %, the nickel concentration considered in this work) approach or may even meet locally the magnetic instability criteria leading to large fluctuations or even to the formation of local magnetic moments or to small superparamagnetic clusters.

II. EXPERIMENT

We study the AMR of three $Pd_{1-y}Ni_yH_x$ films with $y=6$ at. % and for $T<20$ K. The thicknesses d are, respectively, 100, 50, and 25 Å. These films are charged electrochemically with hydrogen up to a saturated state with $x=x_1\simeq0.95$. Hydrogen is then released step by step in a controlled way leading to concentrations $x_2\simeq0.85$, $x_3\simeq0.75$, and $x_4\simeq0.65$. No experiments are done for $x<0.65$ because one enters then into a two-phase region. At each step, we measure the resistance $R(T)$, the magnetoresistance $\Delta R(T,H)$, and the Hall effect. More details on the preparation are given in Refs. 3 and 4. In all cases we have checked that our films fulfill the condition of 2D dimensionality for the localization phenomena.[3,4] In the present paper we will concentrate our attention mainly on the AMR and on the superconducting properties.

III. RESULTS AND DISCUSSION

A. Control of the magnetic state of the films

This state can be checked in a qualitative way through measurements of the Hall effect. For a normal nonmagnetic film one expects a Hall voltage which varies linearly with the applied magnetic field H and which is essentially temperature independent. For magnetic systems one expects an extraordinary Hall effect, related to the magnetization M; for a paramagnetic (with localized moments) or superparamagnetic situation one expects a temperature-dependent Hall coefficient varying like $a+b/T$; for a truly magnetic (i.e., ferromagnetic) case one expects a strong Hall voltage with a nonlinear or even hysteretic behavior as a function of field H.

A detailed investigation of the Hall effect as a function of x and y will be given in a forthcoming work[9] and we will give here only the main features.

(a) For the saturated ($x=x_1\simeq0.95$) samples we observe that the Hall coefficient is strictly temperature independent between 77 and 1.5 K for all thicknesses. The behavior is thus that of a normal nonmagnetic metal. But it does not mean, of course, that the parameter I must be zero. For instance, we also observe a temperature-independent Hall coefficient in pure Pd films ($x=0, y=0$) where it is known that $I\sim0.8$–0.9, i.e., there are magnetic fluctuations but no local moments nor long-range magnetic correlations.

(b) Signs of magnetism appear first in the thickest film ($d=100$ Å) when one decreases x. The Hall coefficient becomes slightly temperature dependent, increasing at low temperatures in the x_2 and x_3 states. Nonlinear behavior appears only in the x_4 state.

(c) For the 50- and 25-Å films the temperature dependence of the Hall coefficient appears only in the x_3 and x_4 states.

One can conclude qualitatively that signs of magnetism appear when x decreases, these signs being largest in the thickest films.

B. AMR in the saturated state ($x=x_1\simeq0.95$)

In Fig. 1 we plot the magnetoresistance ($\Delta R_\square/R_\square^2$) as a function of H (perpendicular to the film) for the $d=25$ Å sample and for different temperatures. The AMR is always positive i.e., we are in the WAL regime due to the large spin-orbit–scattering rate. The solid lines in Fig. 1

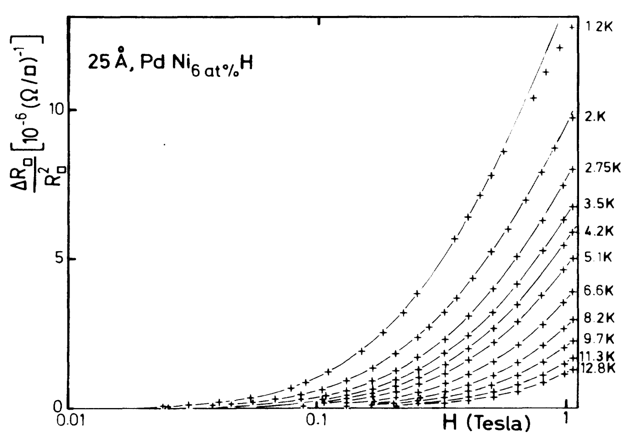

FIG. 1. Anomalous magnetoresistance of a 25-Å film at different temperatures. The curves are best-fitted using the WAL model. The deviations at high fields for the lowest temperature are due to superconductivity.

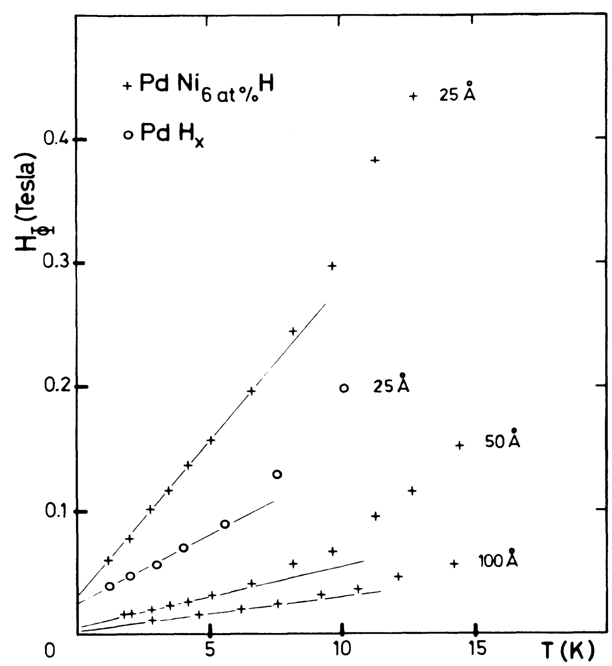

FIG. 2. Phase-breaking field as a function of temperature. The linear dependence at low temperature reflects the e-e interactions. Note also the increase of H_ϕ with Ni content for the 25-Å film.

are theoretical fits using the $\tau_{s.o.}^{-1} \gg \tau_S^{-1}, \tau_{in}^{-1}$ limit, which was also used for the Pd and PdH films.[3,4] The AMR is then given by

$$\frac{\Delta R_\square}{R_\square^2} = \frac{e^2}{2\pi^2\hbar}(\beta-\alpha)Y(H/H_\phi) ,$$
$$Y(x) = \ln x + \psi\left[\frac{1}{2}+\frac{1}{x}\right] . \quad (1)$$

β is the Maki-Thompson contribution[11] and α the WAL one.

$H_\phi = 2H_S + H_{in}(T)$ is the phase-breaking field which comprises the spin-flip field H_S and the inelastic field $H_{in}(T)$. In Fig. 2 we plot the fields $H_\phi(T)$ for the three thicknesses together with the result for a 25-Å Pd-H (without Ni) film. It is quite clear that the analysis used in Refs. 3 and 4 applies here also, i.e., $H_{in}(T) = BT + CT^3$, where the first term results from the inelastic e-e scattering and the second term from the electron-phonon scattering; note that the linear part BT is well marked for Pd-Ni-H. In the following we will concentrate our attention on the values of τ_S^{-1} and τ_{ee}^{-1} extracted from the linear region of $H_\phi(T)$ and related to H_ϕ and H_{ee} through the relation

$$H_i = (\phi_0/4\pi D\tau_i) = (\phi_0/4\pi L_i^2) \quad (2)$$

($i = s$ or ee, ϕ_0 the flux quantum, D the electronic diffusion constant which we deduce from the measurements of the superconducting critical field H_{c2},[3,4] and L_i the corresponding coherence length).

Table I presents the main results concerning Pd-Ni-H and Pd-H.[3,4] The main result from this table concerns τ_{ee}^{-1}, which is systematically higher for Pd-Ni-H compared to PdH. The presence of nickel, in fact, increases τ_{ee}^{-1} by roughly a factor of 2.5. For a more precise comparison it is necessary to normalize the data to the same $R_\square$ value for each film thickness, i.e., to compare $\tau_{ee}^{-1}R_\square^{-1}$. The ratios between Pd-Ni-H and PdH are then, precisely, 3.56 for the 100-Å film, 2.9 for the 50-Å film, and 2.5 for the 25-Å film. These data clearly indicate that the difference between Pd-Ni-H and PdH cannot be ascribed to a variation of $R_\square$.

The theoretical values of τ_{ee}^{-1} have been calculated in two limits: First, a spin-independent contribution resulting mainly from the long-range part of the Coulomb interaction has been estimated in Refs. 12 and 13; it is given by

$$\tau_{ee}^{-1} = (e^2/2\pi\hbar^2)R_\square k_B T \ln(\tau_{ee} k_B T/\hbar) \quad (3)$$

(this expression is slightly different, concerning the logarithmic term, from a preceding one used in Refs. 3 and 4; the justification for this is given in Ref. 13).

A spin-dependent contribution related to the short-range part of the Coulomb interaction and to the electron scattering from paramagnons has been estimated in Refs. 5 and 6; it is given by

$$\tau_{ee}^{-1} = \frac{3e^2}{2\pi\hbar^2} R_\square \frac{I}{1-I} k_B T \ln\left[\frac{\tau_{ee} k_B T}{\hbar(1-I)}\right] \quad (4)$$

[the validity of this formula implies that I is close to 1 and that $(k_B T\tau)/\hbar(1-I) < 1$, where τ is the elastic lifetime; we have checked that this is always the case for our situation].

In Table II we report the experimental values of τ_{ee}^{-1} for Pd-Ni-H compared with the values calculated from the first spin-independent contribution [formula (3)] (all values are again for $T = 1$ K).

It can be seen immediately that the calculated values from formula (3) are always smaller than the observed ones, indicating that there must be a further contribution to τ_{ee}^{-1}.

By adding tentatively the two theoretical contributions (3) and (4) we can calculate the value of I necessary to account for this discrepancy and therefore explain the experimental results. These values are given in the third column on Table II. As formula (4) is derived for extended paramagnons, it can only be applied to Pd-Ni-H in a semiquantitative way. Nevertheless, all the data contained in Tables I and II indicate clearly that the presence of the Ni atoms increases the e-e scattering rate τ_{ee}^{-1} in a way which cannot be related to a variation of $R_\square$, but which must be related to a large value of I. The estimated values of I show also a thickness dependence which agrees qualitatively with the trends discussed above concerning the evolution of the magnetic state of these films: I is largest for the thickest film.

TABLE I. Experimental values of the spin-flip (τ_S^{-1}) and electron-electron (τ_{ee}^{-1}) scattering rates for saturated Pd-Ni-H and PdH films with comparable sheet resistance $R_\square$ and diffusion constant D.

		d (Å) 25	50	100
$R_\square$ (Ω)	Pd-Ni-H	369	85	25
	Pd-H	360	107	33
D (10^{-4} m^2/s^{-1})	Pd-Ni-H	1.6	3.5	5.8
	Pd-H	1.7	3.0	4.8
τ_S^{-1} (10^{10} s^{-1})	Pd-Ni-H	1.4	0.6	0.5
	Pd-H	1.3	2.6	0.8
τ_{ee}^{-1} (10^{10} s^{-1}) at $T=1$ K	Pd-Ni-H	2.5	1.06	0.8
	Pd-H	1.04	0.46	0.3

TABLE II. Comparison between the experimental and calculated [formula (3)] e-e scattering rate at $T=1$ K. Column 3 gives the calculated values of I [formula (4)] needed in order to explain the difference between the first two columns. The last column gives an independent estimate of I from the decrease of the superconducting T_c with thickness (Fig. 3).

d (Å)	τ_{ee}^{-1} (Expt.) (10^{10} s^{-1})	τ_{ee}^{-1} [formula (3)] (10^{10} s^{-1})	I [formula (4)]	(Superconductivity)
25	2.5	0.58	0.61	0.70
50	1.06	0.18	0.67	0.88
100	0.8	0.066	0.81	0.91

Table I also gives the variation of τ_S^{-1} for Pd-Ni-H and PdH; here the situation is somewhat less clear, because for $d=25$ Å there is a slight increase of τ_S^{-1} with Ni while there is a decrease for $d=50$ and 100 Å. But this is not too surprising if one attributes τ_S^{-1} to spin-flip scattering from localized moments due to the presence of a small concentration of d impurities (such as Fe, Co, . . .). A qualitative calculation given in Ref. 3 showed that the value of τ_S^{-1} can be accounted for by only a few ppm of such impurities and it is, of course, difficult to ascertain that this impurity content is constant from one film to the other. To first order one can say that Ni does not contribute to elastic spin-flip scattering, in agreement with the idea that there are no localized moments on the nickel atoms (because $I<1$), but only inelastic fluctuations (paramagnons).

C. Superconductivity

It is known that the addition of Ni decreases the superconducting T_c of bulk PdH, with T_c going towards zero for $y \simeq 15$ at. %.[14,15] A detailed experimental analysis of the electron-phonon coupling in this system showed that only part of the decrease of T_c can be attributed to modifications of the electronic and atomic structure of the host metal by Ni. It is then interesting in this context to see whether it can be related to the above-discussed magnetic fluctuations. In Fig. 3 we plot T_c as a function of $R_\square$ for saturated PdH and Pd-Ni-H. A decrease is observed in both cases, but it is far stronger (about 4 times) in Pd-Ni-H, indicative of a specific effect of the Ni atoms. The decrease of T_c in PdH films can be attributed to Anderson localization in relation with their nonmagnetic atomic disorder measured by $R_\square$; this situation has been analyzed in detail[16] and it can be expressed as

$$\ln(T_c/T_{c0}) = -A_2 R_\square \tag{5}$$

(T_{c0} is the critical temperature of the bulk system and A_2 is a parameter function of $\ln T_c\tau$, where τ is the elastic scattering lifetime).

One can recall here that the decrease of T_c is related to the increase of the Coulomb repulsion with disorder, to modifications of the retardation effects, as well as to a change of the Fermi-level density of states.

The measured value of A_2 (Fig. 3) for PdH is equal to 2 (if $R_\square$ is expressed in kΩ), which is of the same order of magnitude as the theoretical estimates[16] or the values observed in other systems.[17,18]

The situation with paramagnon-like fluctuations has been analyzed by Berk and Schrieffer[19] in clean 3D systems and in Refs. 20 and 21 for disordered films. The latter leads to a further decrease of T_c given by

$$\ln\left[\frac{T_c}{T_{c0}}\right] = -\frac{R_\square}{2}\frac{I}{1-I}\frac{e^2}{2\pi^2\hbar}\ln^3\left[\frac{\hbar}{\pi k_B T_c \tau}\right] \tag{6}$$

(this formula is valid for I close to 1 and $k_B T_c\tau/\hbar \ll 1-I$).

Adding again both contributions (5) and (6), we obtain a new estimate of I given in Table II (last column). One can see that this second estimate gives values somewhat larger than the ones deduced from τ_{ee}^{-1}, but one has to consider that we neglect the effect of τ_S^{-1} on T_c, whose consideration would decrease the values of I somewhat. It is to be noted also that all the estimates of I are perhaps systematically too large because the theoretical models consider only extended paramagnons, while we have perhaps more localized paramagnons, which are known to modify more strongly τ_{ee}^{-1} and probably also T_c for a given value of I. Nevertheless, this description in terms of I gives a good understanding of the much more rapid T_c

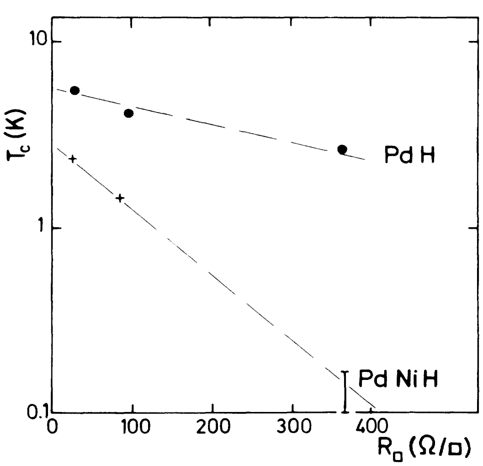

FIG. 3. Variation of the superconducting T_c as a function of $R_\square$. The lowest point for Pd-Ni-H has been inferred from the measurement of the Maki-Thompson coefficient β (see Ref. 4).

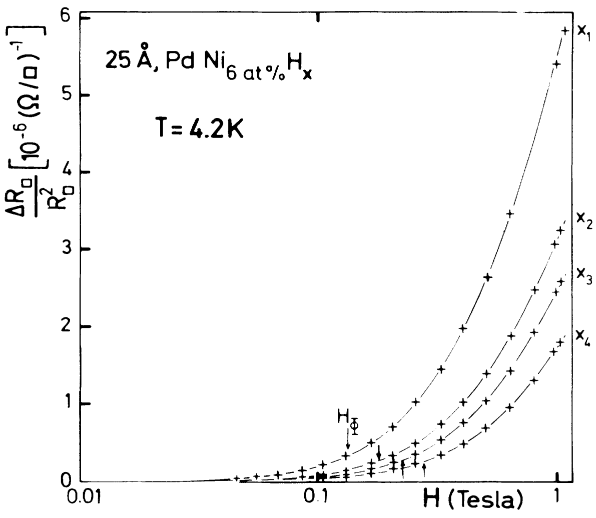

FIG. 4. Magnetoresistance at a given temperature and thickness, as a function of x varying from $x_1 \sim 0.95$ to $x_4 \sim 0.65$. The arrows indicate the values of H_ϕ for each concentration.

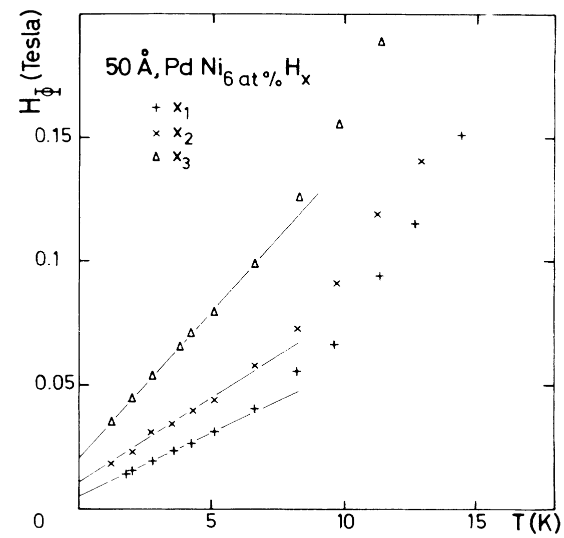

FIG. 5. Phase-breaking field as a function of T for different x values. Note the increase of H_S (at $T=0$) and of the linear slope as x decreases from $x_1 \sim 0.95$ to $x_3 \sim 0.75$.

decrease with $R_\square$ in Pd-Ni-H than in Pd-H films; the decrease of T_c in bulk Pd-Ni-H is probably also related to large *e-e* effects.

D. Variation of τ_{ee}^{-1} and τ_S^{-1} with x

In Fig. 4 we plot the AMR for given thickness and temperature, but different x values (with $x < x_1$). One can note that $\Delta R_\square / R_\square^2$ becomes systematically smaller, which means that the phase-breaking field $H_\phi(T)$ increases as x decreases. This is shown in Fig. 5, where it appears that both H_S (the spin-flip field) and H_{in} (the inelastic field) are enhanced. Table III summarizes most of the data.

In the preceding section we attributed τ_S^{-1} mainly to the presence of magnetic impurities; but the strong and systematic increase of τ_S^{-1} when x decreases cannot be attributed to these impurities, their concentration being independent of x. This suggests instead that a small fraction of the Ni atoms meets the magnetic instability criteria, leading to the formation of localized moments (on isolated Ni atoms or on small clusters) which then give rise to spin-flip scattering; these moments can also explain the progressive appearance of an anomalous T-dependent Hall effect. On the other hand, the systematic increase of τ_{ee}^{-1} can then be attributed to all the other Ni atoms which have approached but not met the magnetic instability criteria, i.e., we attribute it to an increase of I whose value is calculated using the same procedure as before (see Table III); in any case, it cannot be accounted for by an increase of $R_\square$, whose variation is, by far, too small to explain the variation of τ_{ee}^{-1}.

It is to be noted also that the superconducting T_c disappears (i.e., $T_c \ll 1$ K) for all nonsaturated samples, an effect which is perfectly in line with the observed increase of τ_{ee}^{-1} and τ_S^{-1}. It is important to remark finally that the increases of τ_S^{-1} and τ_{ee}^{-1} are somewhat thickness dependent, being largest for the thickest film. It means that the tendency towards magnetism is less pronounced in the thinnest films but one must note also that the thinnest

TABLE III. Experimental variation of the spin-flip (τ_S^{-1}) and *e-e* (τ_{ee}^{-1}) scattering rates as a function of decreasing hydrogen concentration; these variations are much stronger than the corresponding variation of $R_\square$. The last column for each thickness gives the value of I estimated using the same procedure as in Table II (column 3). τ_{ee}^{-1} is given at $T=1$ K.

	$d=25$ Å				$d=50$ Å				$d=100$ Å			
x	$R_\square$ (Ω)	τ_S^{-1} (10^{10} s^{-1})	τ_{ee}^{-1} (10^{10} s^{-1})	I	$R_\square$ (Ω)	τ_S^{-1} (10^{10} s^{-1})	τ_{ee}^{-1} (10^{10} s^{-1})	I	$R_\square$ (Ω)	τ_S^{-1} (10^{10} s^{-1})	τ_{ee}^{-1} (10^{10} s^{-1})	I
x_1	369	1.4	2.5	0.61	85	0.6	1.06	0.67	25	0.5	0.8	0.81
x_2	375	2.4	2.8	0.65	95	1.04	1.24	0.70	32	1.47	0.9	0.82
x_3	378	2.9	3.25	0.69	100	2.05	2.02	0.79	35	2.3	1.64	0.87
x_4	374	3.9	3.8	0.72								

films are also the ones which present the largest atomic disorder. This raises the difficult question concerning the relation between magnetism, atomic disorder, and dimensionality. On one hand, it is known from thermodynamics that a low dimensionality disfavors the magnetic state, in agreement with our observations concerning Pd-Ni-H; a similar observation also holds for the nonhydrogenated Pd-Ni alloys, where we observe a systematic decrease of the ferromagnetic T_c as the thickness d decreases.[22] On the other hand, the effect of atomic disorder on magnetism has been discussed extensively by Fukuyama[23] and Béal-Monod;[24-26] increasing the disorder is shown to lead to an increase of the static[23] and q-dependent[24] Stoner factor. This should thus favor the occurrence of magnetism in the thinnest films, but it is argued also[25,26] that a too drastic increase of the disorder can lead to a reversed effect as the extended paramagnons tend to become localized, damping thus all kinds of long-range magnetic correlations. For our case it is somewhat difficult at this stage to ascertain whether the tendency towards magnetism is dominated by dimensionality or by disorder effects.

IV. CONCLUSION

From the analysis of the AMR we mainly show that the presence of a small Ni concentration increases drastically the inelastic electron-electron scattering rate in a way which is not related to atomic disorder or to the resistance per square $R_\square$. We propose that this increase of τ_{ee}^{-1}, which corresponds also to a decrease of the coherence length, is, in fact, due to an increase of the Stoner factor $S=1/(1-I)$; the decrease and disappearance of the superconducting T_c with Ni content (or by decreasing the hydrogen concentration x) also strongly supports this idea. At the same time, we observe an increase of the elastic spin-flip scattering rate τ_S^{-1}, especially for the low-x values and we attribute it to a fraction of localized spins due either to the presence of some magnetic impurities or to some Ni atoms or clusters which have crossed the magnetic phase boundary $I>1$. We observe also that all these effects are thickness dependent being largest for the thickest film. This indicates that the tendency towards magnetism is less pronounced either when the thickness decreases or when the atomic disorder measured by $R_\square$ increases.

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