Preprint

STRAIN DEPENDENCE OF EXCHANGE INTERACTIONS IN DILUTE PdFe ALLOYS AND IN PURE Pd

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We have measured the magnetostriction and the pressure dependence of the Curie temperature[1] in dilute ferromagnetic alloys of Fe in Pd, and find that the magnetization decreases with volume, whereas the Curie temperature increases with volume. Theoretical considerations show that these results are consistent and that each provides independent evidence of a positive strain-dependence of the "s-d" exchange interaction between the conduction electrons and the magnetic moments associated with the Fe atom. In our analysis we employ the experimental value of the magnetostriction in pure Pd.[2] The relatively small value of the latter indicates that the exchange interaction responsible for the enhancement of the spin susceptibility in paramagnetic Pd has a negative strain-dependence, like the exchange interaction in ferromagnetic Ni,[3] and in contrast with the s-d exchange interaction in the dilute ferromagnetic PdFe alloys.

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ABSTRACT

Measurements of the magnetostriction and the pressure dependence of the Curie temperature in ferromagnetic alloys of Pd containing 0.3, 1 and 3 atomic percent Fe provide independent evidence for a positive strain-dependence of the exchange interaction between the conduction electrons and the local moments, which contrasts with the negative strain-dependence of the exchange interaction between the conduction electrons in pure Pd.

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Doniach and Wohlfarth[4] evaluate the saturation moment μ per Fe atom at zero temperature in a dilute ferromagnetic alloy of Fe in Pd. They show that the giant moment μ is

$$\mu = \mu_o(1+J\chi) \,, \tag{1}$$

where J is a normalized exchange interaction parameter and χ is the atomic susceptibility of the Pd matrix. Thus the volume derivative of μ is

$$\frac{\partial \ln\mu}{\partial \ln V} = \frac{J}{1+J} \left(\frac{\partial \ln J}{\partial \ln V} + \frac{\partial \ln \chi}{\partial \ln V}\right) , \tag{2}$$

since the moment μ_o on the Fe atom is assumed to be independent of volume.

The magnetostriction quadratic in field associated with the paramagnetism of pure Pd provides an estimate of $\frac{\partial \ln\chi}{\partial \ln V}$. The volume magnetostriction linear in field of the ferromagnetic PdFe alloys provides an estimate of $\frac{\partial \ln\mu}{\partial \ln V}$ through the thermodynamic relation,

$$\frac{1}{V}\frac{\partial V}{\partial H} = - \frac{1}{\Omega_c}\frac{\partial \mu}{\partial P} = + \frac{\kappa\mu}{\Omega_c}\frac{\partial \ln\mu}{\partial \ln V} , \tag{3}$$

where Ω_c in the volume per Fe atom and κ is the compressibility, which we assume equal to that of pure Pd (5.24×10^{-4} bar^{-1}). The ratio $J\chi/(1+J\chi)$ may be estimated from measurements of the saturation magnetization of the PdFe alloy and we obtain a value of $\partial\ln J/\partial\ln V$ by substitution in Eq. (2).

In the static limit Doniach and Wohlfarth's theory is equivalent to that of Kim,[5] who shows that the Curie temperature of a dilute ferromagnetic alloy varies like

$$T_c \propto J^2 \chi \tag{4}$$

The volume derivative of T_c is therefore

$$\frac{\partial \ln T_c}{\partial \ln V} = \frac{2\partial \ln J}{\partial \ln V} + \frac{\partial \ln \chi}{\partial \ln V} \tag{5}$$

Thus a direct measurement of the pressure dependence of T_c, when combined with the value of $\partial \ln \chi/\partial \ln V$ for pure Pd, provides an independent estimate of $\partial \ln J/\partial \ln V$.

We show in Fig. 1 the longitudinal magnetostriction of pure Pd and of two of the PdFe alloys. We assume the magnetostriction to be isotropic and obtain the volume derivative of the susceptibility of pure Pd by substituting $V^{-1}\partial V/\partial H = 3\ell^{-1}\partial \ell/\partial H$ into the thermodynamic relation,

$$\frac{1}{V}\frac{\partial V}{\partial H} = -\frac{K\chi}{\Omega}\frac{\partial \ln \chi}{\partial \ln V}\cdot H \ , \tag{6}$$

Ω being the atomic volume. In accordance with Eq. (6), the measured magnetostriction of pure Pd integrated from zero field as shown in Fig. 1 is quadratic in the field H, yielding a value[2], $\partial \ln \chi/\partial \ln V = -3.3 \pm 0.1$.

The magnetostriction of $Pd_{97}Fe_{03}$ is linear in H above H ~ 5 kOe, in accordance with Eq. (3) when μ is assumed independent of H. The slope of this linear variation, together with the measured saturation magnetization, gives $\partial \ln\mu/\partial \ln V = -0.06 \pm 0.01$. The non-linear magnetostriction below H ~ 5 kOe is characteristic of these alloys in the field and temperature range where their magnetization is also appreciably field-dependent.[6] In sample $Pd_{99.7}Fe_{00.3}$ this non-linear field-dependence persists up to the highest fields at 4.2°K, since this is close to the Curie temperature of the alloy. However at 1.7°K a satisfactory linear variation over a wide field range is obtained as shown in Fig. 1.

The resultant values of $\partial \ln\mu/\partial \ln V$ for these alloys and also for $Pd_{99}Fe_{01}$, whose magnetostriction is not shown in Fig. 1, are listed in Table 1. The values of $J\chi/(1+J\chi)$ for substitution in Eq. (2) were obtained from the measured saturation magnetization for each alloy by taking $\mu_o = 3.3\ \mu_B$ for the local moment on an Fe site measured by large-angle neutron scattering.[7]

The resistivity of the most dilute PdFe alloy has a discontinuity in the temperature dependence near the Curie temperature T_c, as shown in Fig. 2. Curves obtained at atmospheric pressure and at high pressure in different apparatus[8] show that the discontinuity associated with T_c decreases at a rate $dT_c/dP = -4 \pm 4\times10^{-6}$ °K bar^{-1}. The corresponding value of $d\ln T_c/d\ln V$ is given in Table 1, and $d\ln J/d\ln V$ is evaluated by use of Eq. (5).

We attribute the linear increase of the resistance with temperature below T_c to the increasing spin-disorder scattering. The curves of Fig. 2 show that both the slope of the linear region and the intercept extrapolated to absolute zero increase with pressure, which means that both the spin-disorder and the residual scattering increase with pressure. DeGennes[9] has estimated the spin-disorder scattering in rare-earth metals, and shows that the resistivity is proportional to the square of the exchange interaction between the conduction electrons and the localized 4f-states. In the PdFe alloys, the spin-disorder scattering is influenced by the enhancement effects of the Pd host. However different matrix elements of both the exchange energy and the "enhancement operator" enter into this process from those which determine T_c and μ. Therefore information on the spin-disorder scattering does not lead to any simple conclusions concerning the strain dependence of J.

The pressure dependence of the Curie temperature of $Pd_{97}Fe_{03}$ was measured by an ac method in which the sample forms the core of a small transformer and the change in the relative initial susceptibility with temperature is recorded at different pressures.[8] Typical curves are shown in Fig. 3. It is difficult to define T_c unambiguously, but it is clear that the curves shift to lower temperatures with increasing pressure. Defining T_c by extrapolating the rapidly rising part of each curve to the background, we obtain

$dT_c/dP = -100 \pm 40\times10^{-6}$ °K bar^{-1}, which results in the values of $\partial \ln T_c/\partial \ln V$ and $\partial \ln J/\partial \ln V$ given in Table 1.[10]

The different measurements yield values of $\partial \ln J/\partial \ln V$ which are always positive and average to $+2.4 \pm 0.5$.[11] It seems that the positive strain-dependence of the s-d exchange interaction approximately cancels the decrease of the susceptibility of the Pd matrix with increasing volume. This results in a small negative magnetostriction (since $\mu \sim J\chi$ from Eq. (1)) and positive strain-dependence of T_c (since $T_c \sim J^2\chi$ from Eq. (4)). It is interesting to note that in pure Pd the negative strain-dependence of the exchange interaction between the itinerant carriers approximately cancels the increase of the density of states with increasing volume, so that the magnetostriction is relatively small and negative as in the PdFe alloys, but for a quite different reason.[12]

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REFERENCES

1. D. B. McWhan and M. P. Sarachik, Bull. Am. Phys. Soc. 12, 504 (1967).
2. E. Fawcett and G. K. White, J. Appl. Phys. 39, 576 (1968). The measurements reported in this reference were on a sample of Pd which was later found to contain several hundred ppm Fe impurity, and which exhibited a strongly temperature and field-dependent susceptibility. The present sample of Pd was made from Matthey-Bishop wire containing 4 ppm Fe and < 10 ppm total metallic impurities, and its susceptibility was field-independent and essentially independent of temperature at low temperatures. We are indebted to E. Bucher for these susceptibility measurements.
3. N. D. Lang and H. Ehrenreich, J. Appl. Phys. 38, 1316 (1967); Phys. Rev. (to be published).
4. S. Doniach and E. P. Wohlfarth, Proc. Roy. Soc. A296, 442 (1967).
5. D. J. Kim, Phys. Rev. 149, 434 (1966).
6. Similar non-linear magnetostriction is observed even at temperatures well below the Curie temperature in an alloy $Pt_{99}Fe_{01}$ where the magnetostriction varies as the square of the magnetization (E. Fawcett and R. C. Sherwood, to be published), in accordance with the prediction of a Stoner model for a weak itinerant ferromagnet (P. Wohlfarth, to be published).

7. J. W. Cable, E. O. Wollan, and W. C. Koehler, Phys. Rev. 138A, 755 (1965); G. G. Low and T. M. Holden, Proc. Phys. Soc. 89, 119 (1966).

8. The experimental methods for the high pressure measurements have been described in previous publications; D. B. McWhan and A. L. Stevens, Phys. Rev. 154, 438 (1967); 139, A682 (1965); D. N. Lyon D. B. McWhan and A. L. Stevens, Rev. Sci. Instrum. 38, 1234 (1967

9. P. G. de Gennes, Compt. Rend. 247, 1836 (1958).

10. Similar measurements were made on an alloy $Pd_{97}Co_3$ and gave $\partial T_c/\partial p = 0.0 \pm 0.1\ °K \times 10^{-3}\ bar^{-1}$, which corresponds to $\partial \ell nJ/\partial \ell nV = +1.7 \pm 0.7$.

11. The decrease of $\partial \ell n\mu/\partial \ell nV$ for $Pd_{97}Fe_{03}$ when compared with the other two alloys suggests a concentration dependence of the magnetostriction. This is to be expected since in this alloy the probability of an Fe atom having another Fe atom nearest neighbor is 30%, whereas in the most dilute alloy it is only 3%. If we exclude the data for $Pd_{97}Fe_{03}$ from the average we obtain the result for dilute PdFe alloys, $\partial \ell nJ/\partial \ell nV = +2.0 \pm 0.2$.

12. This behavior in Pd is analogous to that in ferromagnetic Ni, where Lang and Ehrenreich (Ref. 3) estimate that the negative strain-dependence of the intra-atomic Coulomb repulsion just outweighs the increase of the density of states with increasing volume, resulting in an increase of Curie temperature with pressure.

TABLE 1

Magnetostriction and Pressure Dependence of T_c for PdFe Alloys.

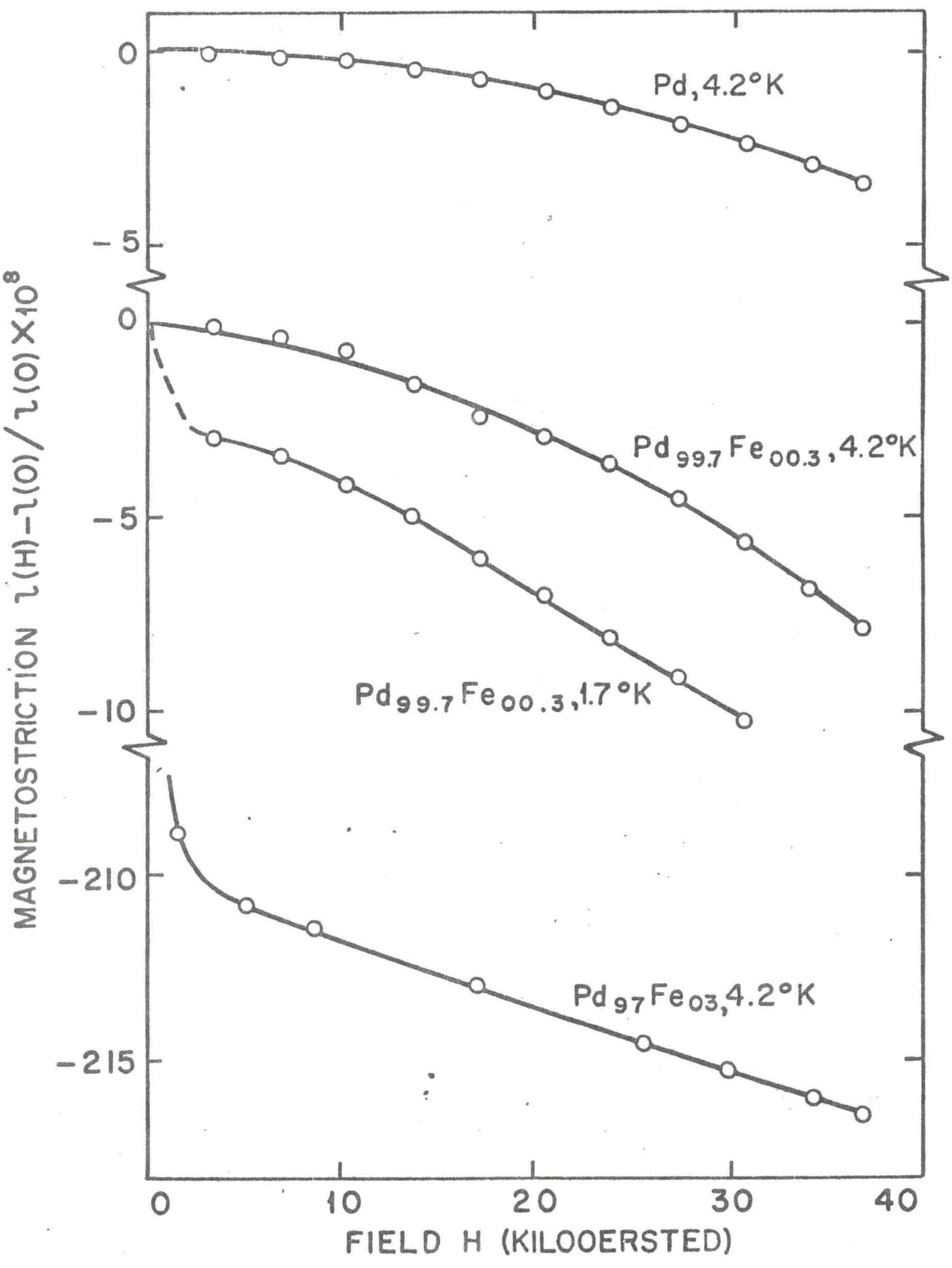
alloy	magnetostriction		pressure dependence	
	$\frac{\partial \ln \mu}{\partial \ln V}$	$\frac{\partial \ln J}{\partial \ln V}$	$\frac{\partial \ln T_c}{\partial \ln V}$	$\frac{\partial \ln J}{\partial \ln V}$
$Pd_{99.7}Fe_{00.3}$	-0.89 ±0.03	+2.0 ±0.1	+1.7 ±1.7	+2.6 ±1.0
$Pd_{99}Fe_{01}$	-1.05 ±0.03	+1.7 ±0.1	-	-
$Pd_{97}Fe_{03}$	-0.06 ±0.01	+3.2 ±0.1	+1.5 ±0.6	+2.5 ±0.4

FIGURE CAPTIONS

Fig. 1 Magnetostriction of pure Pd and PdFe alloys.

Fig. 2 Resistance of an alloy $Pd_{99.7}Fe_{00.3}$ as a function of temperature at atmospheric pressure and at intervals of 15 k bar in pressure up to 45 k bar.

Fig. 3 Change of initial susceptibility vs. temperature for $Pd_{97}Fe_{03}$ at different pressures.



MAGNETOSTRICTION $l(H)-l(0)/l(0)\times10^8$
0
-5
0
-5
-10
-210
-215
Pd, 4.2°K
$Pd_{99.7}Fe_{00.3}$, 4.2°K
$Pd_{99.7}Fe_{00.3}$, 1.7°K
$Pd_{97}Fe_{03}$, 4.2°K
0
10
20
30
40
FIELD H (KILOOERSTED)

Fig. 1

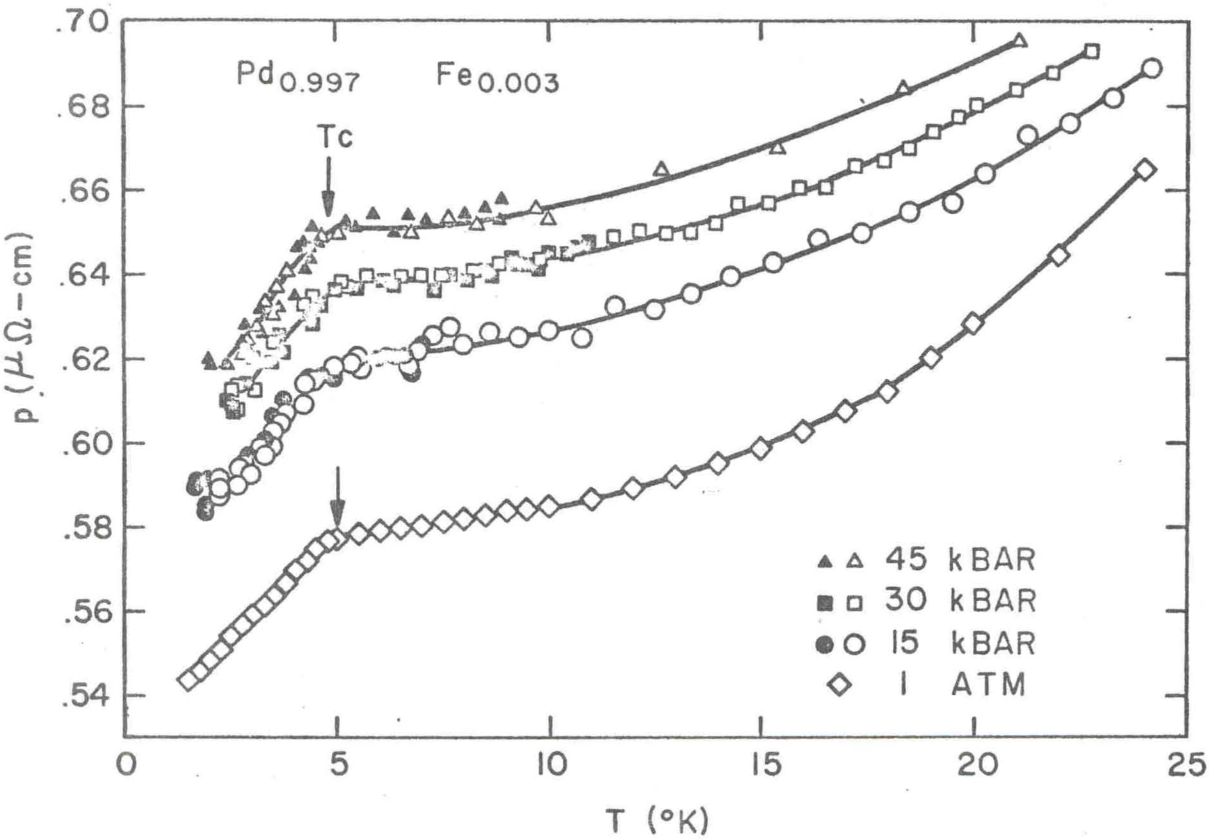
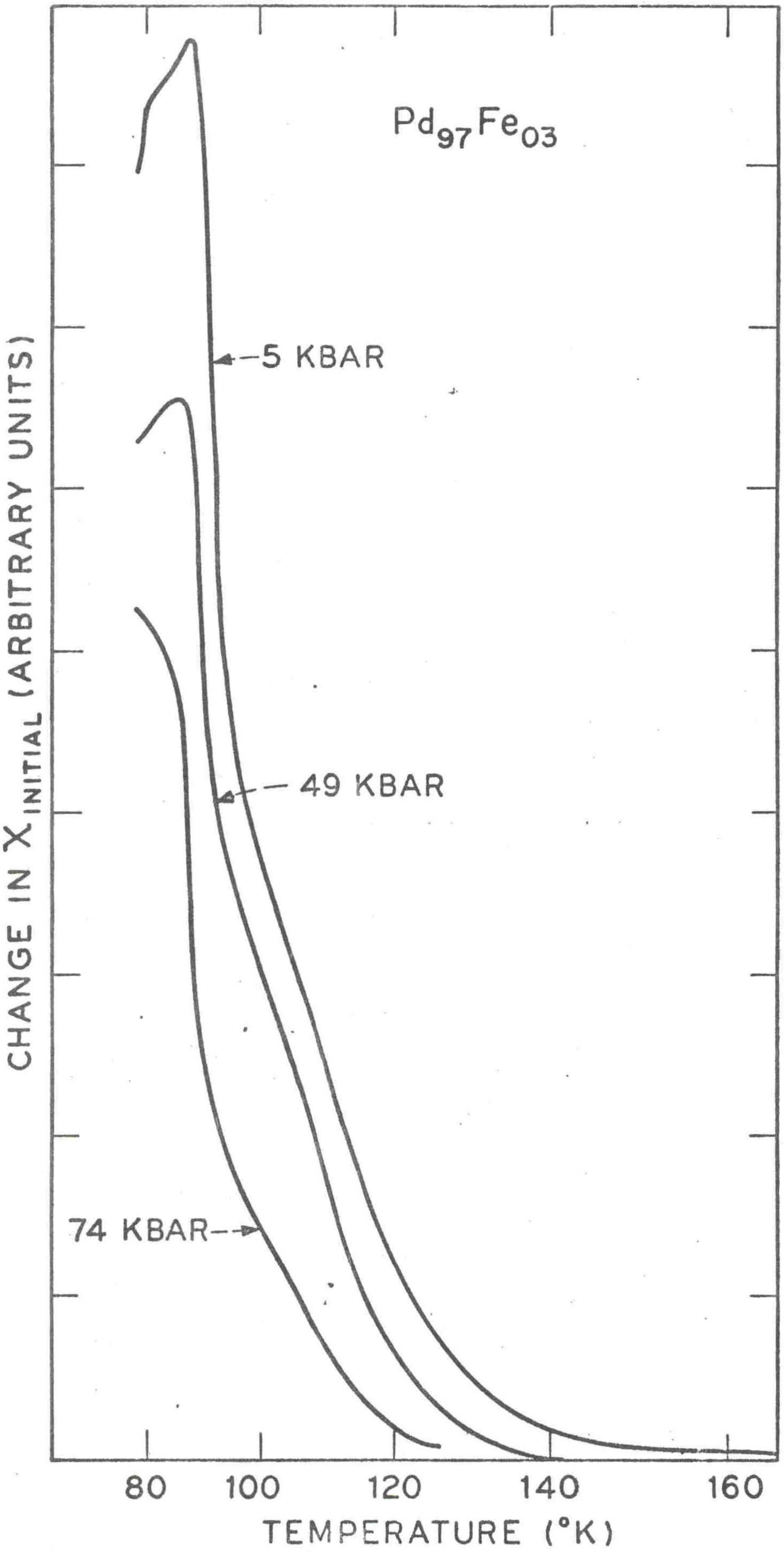

Pd0.997 Fe0.003
Tc
ρ (μΩ-cm)
.70
.68
.66
.64
.62
.60
.58
.56
.54
0
5
10
15
20
25
T (°K)
45 kBAR
30 kBAR
15 kBAR
1 ATM

Fig. 2



$Pd_{97}Fe_{03}$
5 KBAR
49 KBAR
74 KBAR
CHANGE IN $X_{INITIAL}$ (ARBITRARY UNITS)
80
100
120
140
160
TEMPERATURE (°K)

Fig. 3