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# Suppression of the non-linear susceptibilities of ferromagnetic PdFe and PdMn

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**Abstract.** A detailed experimental investigation of the linear and non-linear AC susceptibilities of two ferromagnetic samples, namely PdMn with 0.75 at.% Mn and PdFe with 0.17 at.% Fe, has been carried out as a function of temperature, frequency and amplitude of the AC driving field. In order to explain this behaviour, we have also developed a very simple theory for ferromagnetics, based upon the mean-field model. For these samples the expected divergence of the higher-order susceptibility  $\tilde{\chi}_{nl}$  at  $T_c$  does not occur; instead the measured response tends to zero. According to this theory, the correction term for demagnetization effects seems to outweigh the critical divergence in  $\tilde{\chi}_{nl}$  at  $T \rightarrow T_c$ . The extreme sensitivity of  $\tilde{\chi}_{nl}$  to demagnetization correction is not generally recognized in the recent literature. To obtain the transition temperatures and the critical exponent  $\gamma$  of the samples, the well known Kouvel–Fisher method is used. We have found  $T_c = 1.95 \pm 0.02$  K for PdFe and  $T_c = 2.65 \pm 0.02$  K for PdMn. For PdMn and PdFe, the values of  $\gamma$  are calculated to be  $1.41 \pm 0.05$  and  $1.55 \pm 0.05$ , respectively.

## 1. Introduction

In the last decade, non-linear AC susceptibility measurements have been performed on various kinds of magnetic systems to elucidate whether the peculiar characteristics of these systems testify to an equilibrium phase transition. The non-linear part ( $\tilde{\chi}_{nl}$ ) of the susceptibility has played a decisive role to determine the nature of the phase transition at the critical temperature  $T_c$ . Experimental evidence for a phase transition is based on the observation of a divergence of  $\tilde{\chi}_{nl}$  at  $T_c$ .

For this purpose, we have carried out a series of linear and non-linear AC susceptibility measurements on spin-glass, ferro- and antiferromagnetic samples. The results related to the spin-glass samples and the antiferromagnets have already been published [1–4]. In this paper, we wish to exhibit the results related to some ferromagnetic samples, namely PdMn with 0.75 at.% Mn and PdFe with 0.17 at.% Fe.

The general magnetic properties of PdMn alloys are presented, in detail, in our previous paper [1]. Now, we briefly mention the magnetic properties of PdFe alloys.

Two striking features have been experimentally observed for these alloys: ferromagnetism exists down to 0.01 at.% Fe [5, 6]; and the magnitude of the magnetic moment associated with the Fe atom is very large, about  $10\mu_B$  per Fe atom. However, the magnitude of the moment decreases with increasing concentration. The observed  $10\mu_B$  is much larger than the maximum moment ( $\simeq 2.2\mu_B$ ) per Fe atom according to the Pauli principle. Obviously, part of the observed moment must be due to polarization of the host metal, namely of Pd. Two important facts have become evident from measurements of the Mössbauer effect: first, the giant moment exists not only in the ferromagnetic state but also in the paramagnetic one down to a fairly small amount of Fe, about 30 ppm; secondly,

a spontaneous magnetization exists in the absence of an external magnetic field. For an extensive review, readers are referred to [7] and the references therein.

We present the experimental procedure in section 2, the results and discussion in section 3, and finally our conclusions in section 4.

## 2. Experimental procedure

### 2.1. Theoretical model

According to the mean-field theory of Weiss [8], the magnetization  $M(h, T)$  is given by

$$M(h, T) = M_0 B_s(g\mu_B S H^{\text{eff}}/kT) \quad (1)$$

where  $M_0 = Ng\mu_B S$  and  $B_s$  is the Brillouin function, in which  $H^{\text{eff}} = h + \lambda' M(H, T)$  with  $h$  the applied field and  $\lambda'$  a coupling factor between one spin and its surroundings. We introduce the reduced magnetization  $m$  defined as

$$m = m(h, T) = M(h, T)/M_0. \quad (2)$$

When one absorbs the factor  $g\mu_B S/kT$  into the definition of the Brillouin function and take  $\lambda = M_0\lambda'$ , then

$$m = B(h^{\text{eff}}) = B(h + \lambda m). \quad (3)$$

Note that the temperature dependence of  $m$  is now contained in the coefficients  $b_1, b_2, \dots$  of the expansion

$$m = b_1 h^{\text{eff}} + b_3 (h^{\text{eff}})^3 + \dots \quad (4)$$

or

$$m = b_1 (h + \lambda m) + b_3 (h + \lambda m)^3 + \dots \quad (5)$$

or

$$m(1 - \lambda b_1) = b_1 h + b_3 (h + \lambda m)^3 + \dots \quad (6)$$

It is obvious that according to (6)  $m$  will diverge at a temperature given by  $\lambda b_1 = 1$ . On the other hand, for  $\lambda b_1 \neq 1$  and  $h = 0$ ,  $m$  should be zero. But  $m$  can have a value different from zero, namely spontaneous magnetization can exist. As  $b_1 \propto 1/T$ , the relation  $\lambda b_1 = 1$  gives the critical temperature  $T_c = \theta$  of the Curie-Weiss law. For  $T = T_c$ , the spontaneous magnetization is zero; for  $T < T_c$ ,  $m$  is determined by the expression for  $h = 0$ ;

$$b_1 + b_3 (\lambda m)^2 + \dots = 1/\lambda. \quad (7)$$

Now, let us derive the critical exponents  $\beta$  and  $\delta$  from the relations derived above.

Rewriting (5) for  $T = T_c$  and letting  $b_1 = b_1^c$ ,  $b_3 = b_3^c, \dots$  with  $\lambda = 1/b_1^c$  one finds

$$\begin{aligned} m &= b_1^c (h + \lambda m) + b_3^c (h + \lambda m)^3 + b_5^c (h + \lambda m)^5 + \dots \\ m &= b_1^c h + m + b_3^c (h + m/b_1^c)^3 + b_5^c (h + m/b_1^c)^5 + \dots \\ 0 &= b_1^c h + b_3^c (h + m/b_1^c)^3 + b_5^c (h + m/b_1^c)^5 + \dots \end{aligned}$$

or

$$(h + m/b_1^c) = [-(1/b_3^c)(b_1^c h + b_5^c(h + m/b_1^c)^5 + \dots)]^{1/3}. \tag{8}$$

At  $T = T_c$  the magnetization can be defined as

$$m \propto h^{1/\delta} \tag{9}$$

where  $\delta \geq 1$  [9]. On the other hand in the limit  $h \rightarrow 0$  and  $T = T_c$  equation (8) can be written as

$$m = (b_1^c)^{4/3}(-b_3^c)^{-1/3}h^{1/3}. \tag{10}$$

Comparing (9) and (10) one finds  $\delta = 3$ . One can also see from (10) that  $b_3$  has a negative value.

For  $h = 0$  and  $T < T_c$  to determine the exponent  $\beta$  in the critical expression,

$$m \propto (-\epsilon)^\beta = [(T_c - T)/T_c]^\beta \tag{11}$$

we first write

$$-\epsilon = \frac{1/b_1^c - 1/b_1}{1/b_1^c} = \frac{b_1 - b_1^c}{b_1}. \tag{12}$$

Now (6) is written with  $1 - \lambda b_1^c = 0$  as

$$\begin{aligned} (1 - \lambda b_1) - (1 - \lambda b_1^c) &= b_3 \lambda^3 m^2 + \dots \\ b_1^c - b_1 &= b_3 \lambda^2 m^2 + \dots \end{aligned} \tag{13}$$

or

$$m = (-b_1/b_3)^{1/2} b_1^c (-\epsilon)^{1/2}. \tag{14}$$

By comparing (11) and (14) one finds  $\beta = 1/2$ . The prefactor  $(-b_1/b_3)^{1/2} b_1^c$  is a function of  $T$ , but is almost constant in the  $\epsilon$  region where the result given above is valid.

With the equality  $\gamma = \beta(\delta - 1)$  [9], one finds  $\gamma = 1$ , which can easily be verified from the Curie-Weiss law.

As the last extension of this theory, we propose more general expressions for  $\tilde{X}$  and its derivatives, namely the linear and non-linear susceptibilities of the free spins, as given by, for instance, the Langevin or Brillouin function.

We will use the notation  $d^n m$  for  $d^n m/dh^n$  and  $B', B'', \dots$  for the derivatives of  $B$  with respect to the argument  $(h + \lambda m)$ . The first derivative of (3) is

$$dm = B'(1 + \lambda dm)$$

or

$$dm = B'/(1 - \lambda B'). \tag{15}$$

The relation is valid for all values of  $h$ . For  $h \rightarrow 0$  and  $T > T_c$ , one can obtain the well known Curie–Weiss law. Since  $B' = b_1$  and  $b_1 \propto 1/T$ , (15) is written as

$$dm = b_1/(1 - \lambda b_1). \quad (16)$$

Note that

$$1 + \lambda dm = 1/(1 - \lambda B'). \quad (17)$$

The second derivative of (3) is

$$d^2m = B''(1 + \lambda dm)^2 + B'\lambda d^2m. \quad (18)$$

Using (17) one can decompose (18) as

$$d^2m = [B''/(1 - \lambda B')](1 + \lambda dm)^2 = B''/(1 - \lambda B')^3. \quad (19)$$

This equation is zero at temperatures above  $T_c$  for  $h = 0$ , since  $B'' = 0$ . By virtue of the internal field  $\lambda m$ ,  $B'' \neq 0$  below  $T_c$  even for  $h = 0$ . This means that one should be able to measure a second harmonic response, which is sharply peaked just below  $T_c$ , irrespective of hysteresis and domain-wall movements. Furthermore, it is to be expected that, owing to the critical fluctuations, there will be some second harmonic response too just above  $T_c$ . But these will be suppressed by critical slowing down and demagnetizing effects.

The third derivative of (3) is

$$d^3m = B'''(1 + \lambda dm)^3 + 3B''(1 + \lambda dm)\lambda d^2m + B'\lambda d^3m \quad (20)$$

or

$$d^3m = [B'''/(1 - \lambda B')](1 + \lambda dm)^3 + 3B''[\lambda d^2m/(1 - \lambda B')](1 + \lambda dm).$$

Using (17) in the last expression we find

$$d^3m = B'''/(1 - \lambda B')^4 + 3\lambda(B'')^2/(1 - \lambda B')^5. \quad (21)$$

Since  $B'' = 0$ , for  $h = 0$  and  $T > T_c$ , this equation reduces to

$$d^3m = B'''/(1 - \lambda B')^4. \quad (22)$$

Inserting  $B' = b_1$ ,  $B''' = 6b_3$  in (22) one can find

$$d^3m = 6b_3/(1 - \lambda b_1)^4 = 6[b_3/(b_1)^4](dm)^4. \quad (23)$$

In AC measurements of  $\bar{X}_3$ , the second term in (21) again should lead to some short-range ordering contribution at temperatures below  $T_c$ . For  $T < T_c$  it is not clear at all what is to be expected, as there are at least three contributions involved: two in the expression for  $d^3m$  and one caused by hysteresis effects. Another complicating effect is the enormous influence on the measurements of non-linear susceptibilities by the demagnetizing effect. We have already emphasized the importance of this effect in our previous paper [2] where

the relation between the measured and corrected values of the third harmonic component of the non-linear susceptibility is given by

$$\bar{X}_{3c} = \bar{X}_3 / (1 - D\bar{X}_1)^4 \quad (24)$$

(where  $D$  is the demagnetizing factor) or using the compact notation one obtains

$$\frac{d^3m}{dh_i^3} = \frac{d^3m/dh_e^3}{(1 - D dm/dh_e)^4} \quad (25)$$

where  $i$  and  $e$  subscripts are used to indicate internal and external fields, respectively. Since the corrected linear susceptibility is

$$\frac{dm}{dh_i} = \frac{dm/dh_e}{(1 - D dm/dh_e)} \quad (26)$$

using (26) in (25) one can obtain

$$\frac{d^3m}{dh_i^3} = \frac{d^3m/dh_e^3}{(dm/dh_e)^4} \left( \frac{dm}{dh_i} \right)^4 \quad (27)$$

or reorganizing (27) we obtain

$$\frac{d^3m}{dh_i^3} \left( \frac{dm}{dh_i} \right)^{-4} = \frac{d^3m}{dh_e^3} \left( \frac{dm}{dh_e} \right)^{-4} \quad (28)$$

It is experimentally well known that  $dm/dh$  stays finite but theoretically diverges with the  $-\gamma$  exponent. On the other hand, according to the scaling theory,  $d^3m/dh_i^3$  diverges with the critical exponent  $-(3\gamma + 2\beta)$ . Thus the measured third harmonic will be proportional to

$$d^3m/dh_e^3 = \epsilon^{\gamma-2\beta} (dm/dh_e)^4 \quad (29)$$

For the mean-field theory  $\gamma = 2\beta$  and consequently the measured third harmonic will have a finite value. When  $\gamma > 2\beta$ , as is generally the case, the measured third harmonic will be zero at  $T = T_c$  and consequently will pass through a minimum for some  $T > T_c$ . This will make the actual measurements of the scaling exponents in ferromagnets rather difficult.

To check the above theoretical ideas the linear and non-linear AC susceptibilities of two different ferromagnetic alloys having different impurities, namely PdFe (0.17 at.% Fe) and PdMn (0.75 at.% Mn), have been measured. The results are presented in section 3.

## 2.2. Experimental method

The details of the experimental technique and procedure have already been given in our previous publications [1, 2]. Both samples used were almost spherical in shape and their weights were 0.1083 g for PdFe and 0.2650 g for PdMn. The temperature and frequency ranges of our measurements are 1.2–4.2 K and 15–234 Hz, respectively. The AC field amplitude used is 70 mOe. However, in order to investigate the AC field amplitude dependence of the third harmonic, amplitudes of 0.15, 1.00 and 7.00 Oe were also used.

### 3. Results and discussion

In order to perform an experiment on a ferromagnetic sample, one has to be careful about some points: There are two saturation effects that play an important role in the time-dependent behaviour of magnetization  $M(t)$  in response to a time-dependent field  $H(t) = h \cos(\omega t)$ . These saturation effects are the amplitude of the AC field and the measuring frequency. In the analysis, the third harmonic response is found to be proportional to  $h^3$  [1, 2]. When  $h$  is chosen too large, there is a probability that, in the neighbourhood of  $T_c$ ,  $M$  will reach the saturation value  $M_0$ . In extremum,  $M(t)$  will then be a square wave. Owing to the magnetization effect, the rise-time of the square wave will be bounded by the finite value of the susceptibility,  $X = 1/D$ , giving rise to a trapezoidal or even triangular waveform of  $M(t)$  as a function of time. In any case there will also be a strong frequency dependence. If one uses higher frequency values, one is faced with the 'eddy-current' effect as well as the frequency-dependent saturation in magnetization. As a result the values of  $h$  and  $f$  should be low enough not to affect the susceptibility results. Therefore, we confine ourselves to use a fairly small value of  $h$ , like 70 mOe, and  $f$  below 234 Hz during the experimental process. We should further emphasize that all our susceptibility (linear and non-linear) results are normalized to 1 Oe.

#### 3.1. PdFe (0.17 at.% Fe) alloy

The temperature dependences of the in- and out-of-phase components of the linear susceptibility, for different frequencies, are presented in figures 1 and 2, respectively. As can be seen from figure 1 the in-phase component  $\tilde{X}'_1$  first increases with decreasing  $T$ , and then levels off, leading to saturation at the lowest temperatures. Furthermore, around and below the transition temperature, which will be obtained later on, the response becomes frequency-dependent, increasing with decreasing frequency, and this dependence disappears at the lowest frequencies and reaches its maximum value. The mentioned levelling off is due to the demagnetization effect, which is a general feature of a non-uniform ferromagnet [7].

Comparing figures 1 and 2 one immediately sees that the out-of-phase component  $\tilde{X}''_1$  shown in figure 2 decreases with decreasing frequency, in contrast to the behaviour of  $\tilde{X}'_1$  shown in figure 1. Furthermore, especially at higher frequencies, a maximum appears in  $\tilde{X}''_1$  at temperatures where the levelling-off starts for the corresponding in-phase component in figure 1. These maxima seem to shift to higher temperatures with increasing frequency. The reason for these maxima and their behaviour might be related to the demagnetization effect as well as to incomplete growth of the domains that start to form around the ferromagnetic phase transition temperature of the sample. The growth of the domains means domain-wall movement, which gives rise to energy output, causing an anomaly in the absorption term of the susceptibility.

Here we should mention that the zero-frequency value indicated in figure 1 is obtained by plotting  $\tilde{X}'_1$  ( $\tilde{X}_1$  is not used since it is negligible at the lowest frequency) against the measuring frequencies as a parameter of temperature. The extrapolation of these curves give the zero-frequency values of  $\tilde{X}'_1$ ,  $\tilde{X}_0$ . The saturation value of this, obviously, gives the inverse of the demagnetizing factor  $D$ . In this way  $D$  is found to be 54.

Using the value of  $D$  in (26) we obtained the corrected values of  $\tilde{X}_0^c$ , namely  $\tilde{X}_0^c$ . Near  $T_c$  (transition temperature)  $\tilde{X}_0^c$  varies as [9]

$$\tilde{X}_0^c = A[(T - T_c)/T_c]^{-\gamma} \quad (30)$$

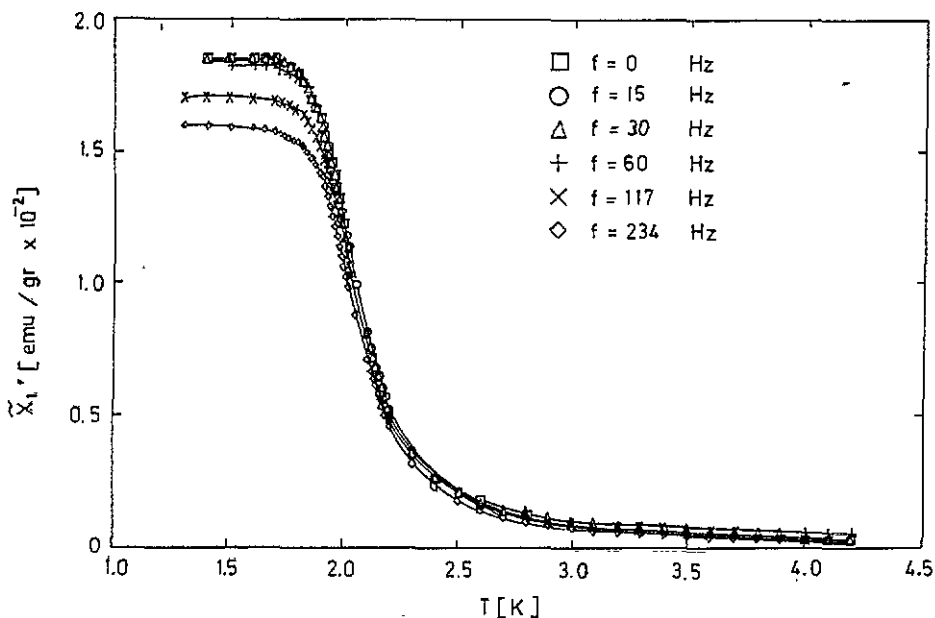


Figure 1. In-phase component of the linear susceptibility of PdFe (with 0.17 at.% Fe) alloy for several frequencies as a function of temperature.

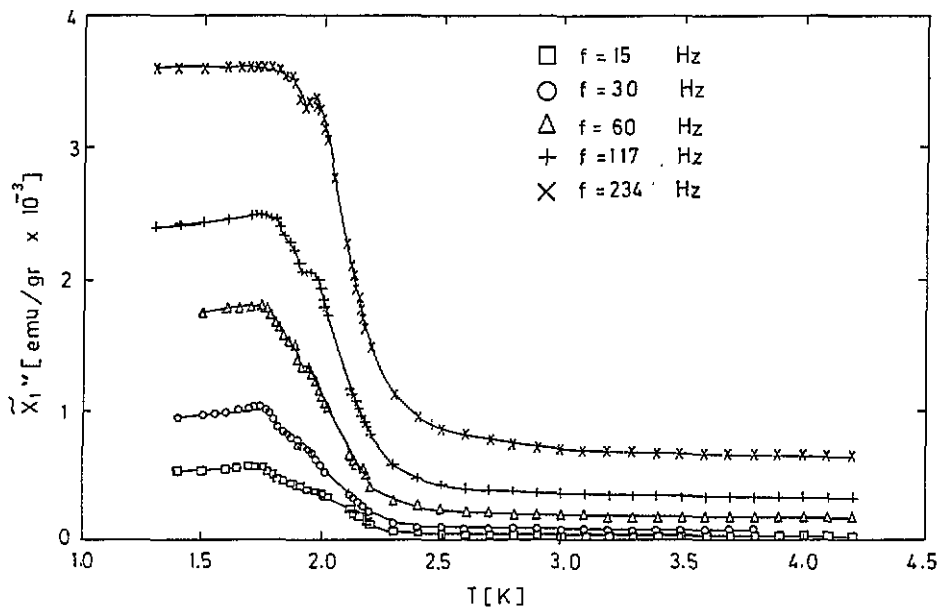


Figure 2. Out-of-phase component of the linear susceptibility of PdFe (with 0.17 at.% Fe) alloy for several frequencies as a function of temperature.

where  $A$  is a constant. By using this relation and applying the well known Kouvel-Fisher method [9], one can determine  $T_c$ , and the critical exponent  $\gamma$ .



For this, one plots

$$\tilde{\chi}_0^c (d\tilde{\chi}_0^c/dT)^{-1}$$

against  $T$ , obtaining a straight line. The slope of this line gives  $\gamma$ , and it intersects the  $T$  axis at  $T_c$ . In order to obtain  $d\tilde{\chi}_0^c/dT$  and the best straight line, we used a three-point numerical differentiation program and linear regression method, respectively. Our data give  $\gamma = 1.55 \pm 0.05$  and  $T_c = 1.95 \pm 0.02$  K.

Another method to obtain  $T_c$  is to extrapolate the slope of  $\tilde{\chi}_0$  at the point where its second derivative with respect to  $T$  is zero. The intersection point of this extrapolation with the saturation (constant) value of  $\tilde{\chi}_0$  gives  $T_c$ . By using this method we obtained almost the same  $T_c$  value as above.

Figures 3 and 4 illustrate the temperature dependence of the in- and out-of-phase components of the third harmonic response, respectively. Both figures indicate that, just above  $T_c$ , the third harmonic of the non-linear susceptibility increases rapidly in a narrow temperature range, passes through a maximum and then tends to zero at  $T_c$ , in agreement with the theory (see section 2). However below  $T_c$ , owing to the dynamical effects, such as hysteresis, relaxation phenomena and remanence, an expected non-linearity is observed.

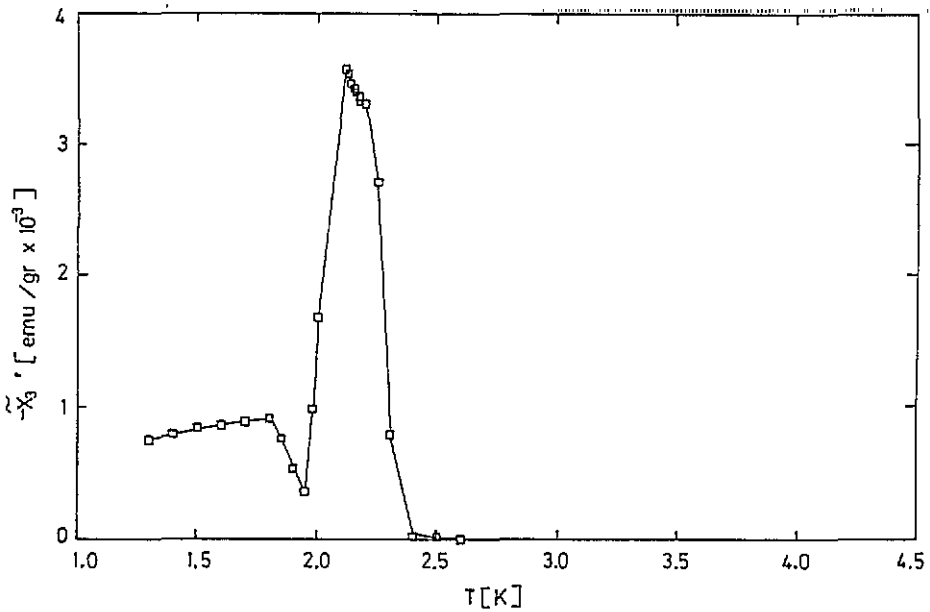


Figure 3. In-phase component of the third harmonic of the non-linear susceptibility of PdFe (with 0.17 at.% Fe) alloy for  $f = 234$  Hz as a function of temperature.

### 3.2. PdMn (0.75 at.% Mn) alloy

In the previous subsection linear and non-linear susceptibility results of a non-uniform ferromagnet have been given and compared with the theory in section 2. Now, we wish to present the results for a 'uniform' ferromagnet, namely PdMn (0.75 at.% Mn). The in- and out-of-phase components of the linear susceptibility appear in figures 5 and 6,

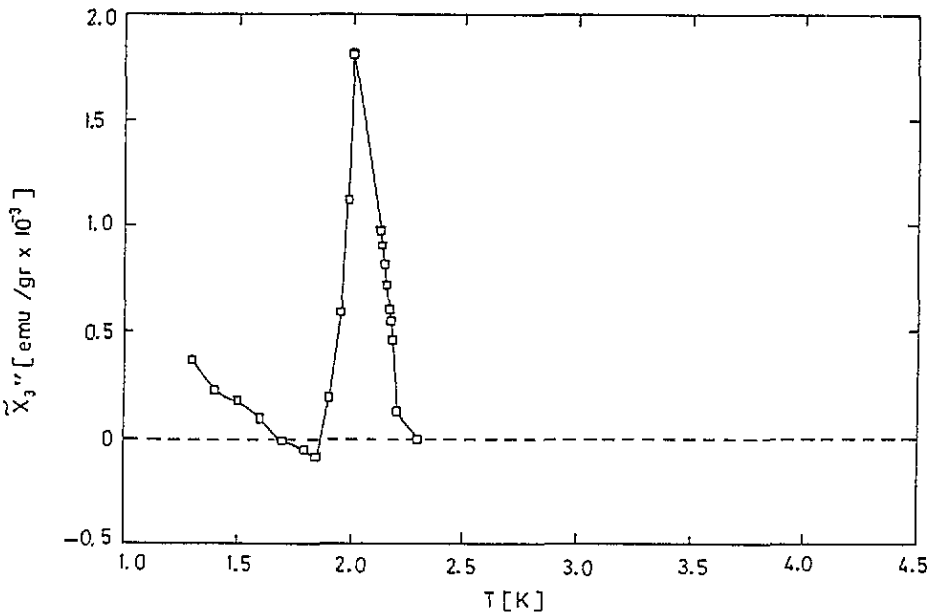


Figure 4. Out-of-phase component of the third harmonic of the non-linear susceptibility of PdFe (with 0.17 at.% Fe) alloy for  $3f = 234$  Hz as a function of temperature.

respectively. As can be seen explicitly from figure 5, the PdMn sample is a very good uniform ferromagnet, since there is a very sharp transition from para- to ferromagnetic phase, not like for the case of PdFe sample discussed in the previous section (compare figures 1 and 5). By looking at figures 2 and 6, one can immediately see that the tiny peaks appearing in figure 2 become well defined and sharp in figure 6, but the behaviour is again the same, i.e. the magnitudes of the peaks increase, and their positions shift to higher temperatures with increasing frequency. However, the dynamical effects below  $T_c$  mentioned for the case of PdFe are not so obvious for the PdMn system.

The zero-frequency value of the linear susceptibility has been found in a similar way as the case of PdFe (see section 3.1), but the result turned out to be almost the same as that of the 15 Hz value. Therefore, for the sake of clarity, the zero-frequency value of  $\tilde{\chi}'_1$  is not shown in figure 5. From the 15 Hz figure the demagnetization factor  $D$  is found to be  $D = 48.9$ .

The ferromagnetic transition temperature  $T_c$  and the critical exponent  $\gamma$  have been calculated in a similar way as for the case of PdFe given in the previous subsection. Our results for  $T_c$  and  $\gamma$  are  $2.65 \pm 0.02$  K and  $1.41 \pm 0.05$ , respectively. Note that the  $\gamma$  value for PdMn is in good agreement with the values predicted in the literature (i.e.  $\gamma = 1.3-1.5$ ), and observed for other ferromagnetic systems [9, 10].

The results for in- and out-of-phase components of the third harmonic  $\tilde{\chi}_3$  are presented in figures 7 and 8. The in-phase component  $\tilde{\chi}'_3$  shown in figure 7 demonstrates that, just above  $T_c$ , a small peak is formed and, at  $T_c$ ,  $\tilde{\chi}'_3$  goes almost to zero. This means that, as the theory in section 2 demonstrates, while approaching  $T_c$  from above,  $\tilde{\chi}'_3$  should diverge, but due to the demagnetizing effect at  $T_c$  this divergence is suppressed forming the mentioned peak. The behaviour of  $\tilde{\chi}'_3$  below  $T_c$  is believed to be due to the dynamical effects. The out-of-phase component  $\tilde{\chi}''_3$  of PdMn is somewhat different from that of PdFe:  $\tilde{\chi}''_3$  approaches

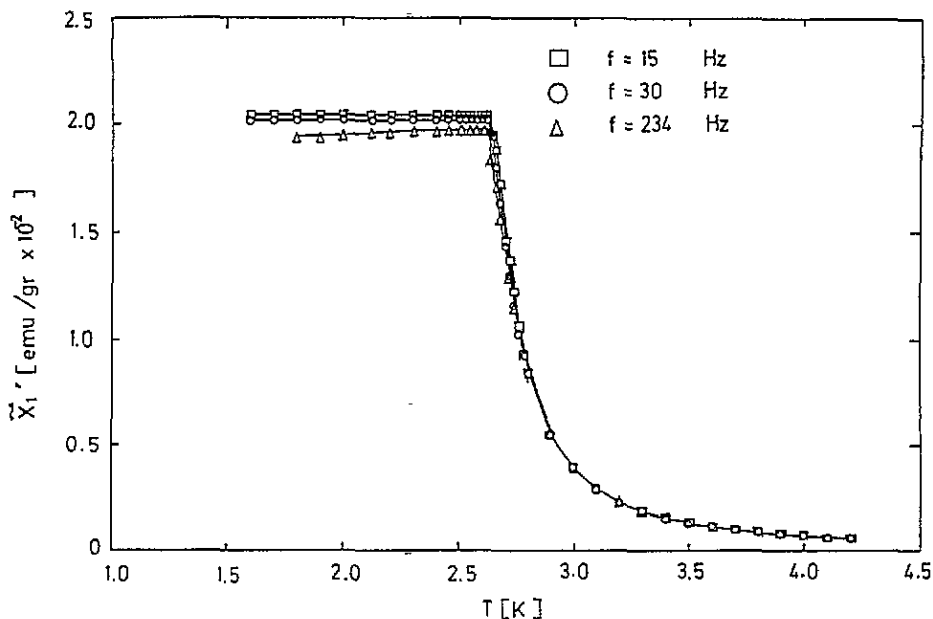


Figure 5. In-phase component of the linear susceptibility of PdMn (with 0.75 at.% Mn) alloy for several frequencies as a function of temperature.

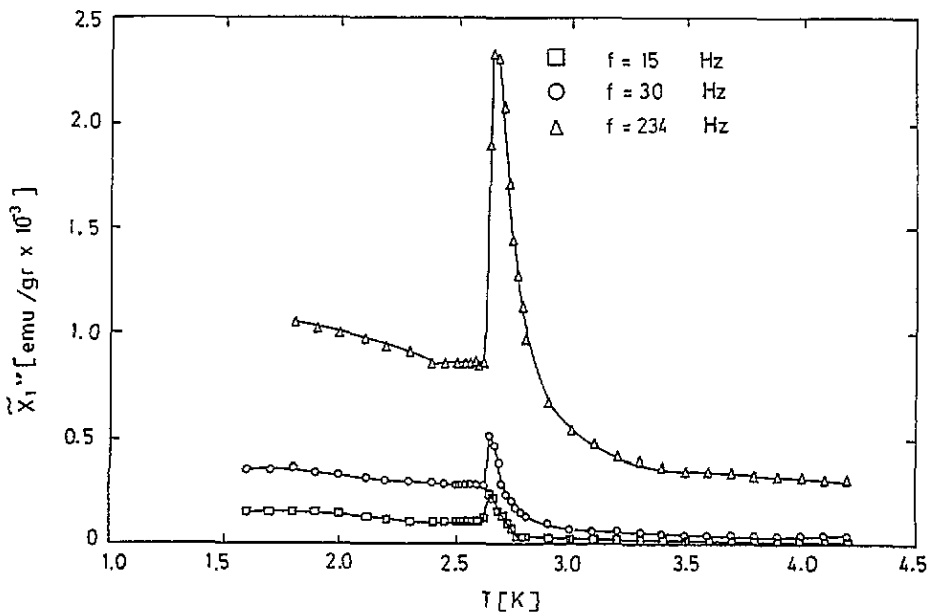


Figure 6. Out-of-phase component of the linear susceptibility of PdMn (with 0.75 at.% Mn) alloy for several frequencies as a function of temperature.

zero at  $T_c$  but then goes to negative values and forms a minimum below  $T_c$ . We attribute this behaviour also to the dynamical effects.

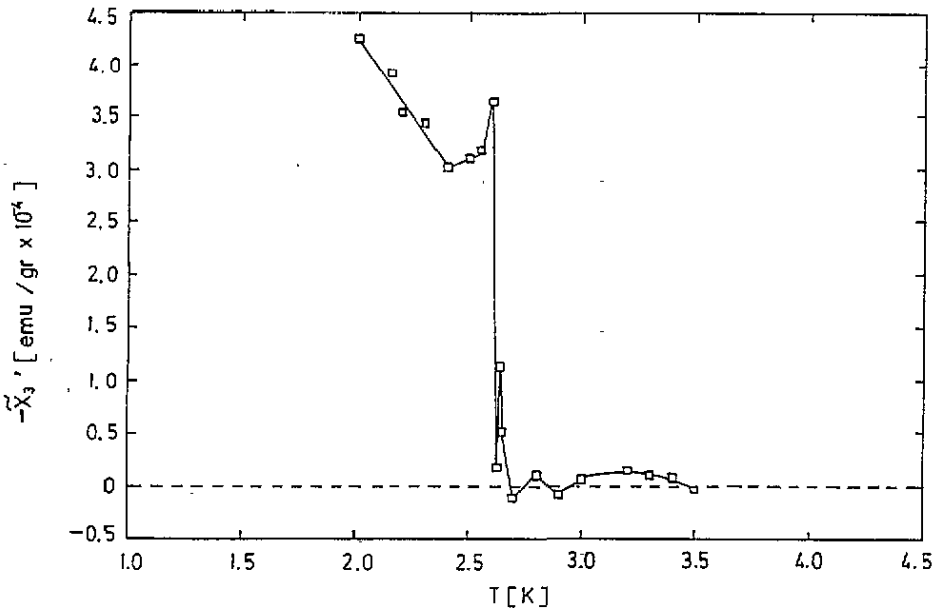


Figure 7. In-phase component of the third harmonic of the non-linear susceptibility of PdMn (with 0.75 at.% Mn) alloy for  $3f = 234$  Hz as a function of temperature.

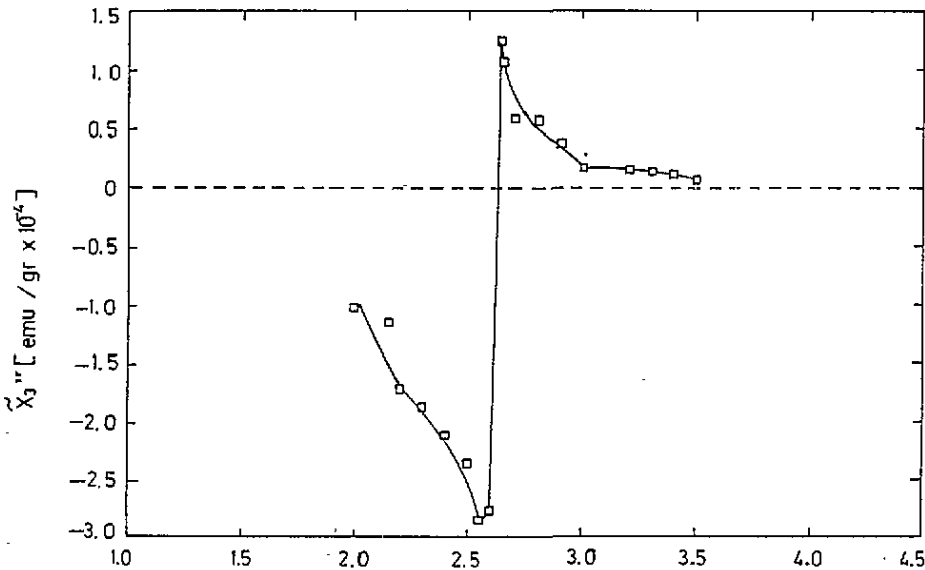


Figure 8. Out-of-phase component of the third harmonic of the non-linear susceptibility of PdMn (with 0.75 at.% Mn) alloy for  $3f = 234$  Hz as a function of temperature.

For the PdMn sample we also investigated the AC field amplitude dependence of the third harmonic. For this we used four different amplitudes (0.07, 0.15, 1.00 and 7.00 Oe). We have found that at 1.00 Oe and above the dynamical effects below  $T_c$  are completely

suppressed, and hence almost no responses for  $\tilde{X}'_3$  and  $\tilde{X}''_3$  are observed.

#### 4. Conclusions

The linear susceptibility measurements on ferromagnetic PdFn and PdMn alloys demonstrate that the former sample is a non-uniform ferromagnet while the latter is a uniform ferromagnet. These observations are in good agreement with the literature [7]. The in-phase and out-of-phase components of the linear susceptibility of ferromagnetic PdMn indicate that below  $T_c$  the in-phase component is almost frequency-independent while the out-of-phase component is frequency-dependent. These observations point out that the measured relaxation time of this sample is almost zero. This can be proven by using the Debye relaxation function. In this conjecture, the measured  $\tilde{X}'_1$  and  $\tilde{X}''_1$  can be approximated as

$$\tilde{X}'_1 \simeq (1/D)[1/(\omega\tau_m + 1/\omega\tau)^2 + 1]$$

and

$$\tilde{X}''_1 \simeq (1/D)[\omega\tau_m/(\omega\tau_m + 1/\omega\tau)^2 + 1]$$

where  $\tau_m$  is the measured and  $\tau$  the intrinsic relaxation time. In general, for ferromagnets  $\tau \rightarrow \infty$  as  $T \rightarrow T_c$ , and is temperature-dependent. Depending on  $\tau_m$  there are three limits for  $\tilde{X}'_1$  and  $\tilde{X}''_1$ :

(i)  $\tau_m \rightarrow \infty$

$$\tilde{X}'_1 \simeq (1/D)(1/\omega^2\tau_m^2) \quad \tilde{X}''_1 \simeq (1/D)(1/\omega\tau_m)$$

(ii)  $\tau_m \rightarrow \text{finite}$

$$\tilde{X}'_1 \simeq (1/D)[1/(1 + \omega^2\tau_m^2)] \quad \tilde{X}''_1 \simeq (1/D)[\omega\tau_m/(1 + \omega^2\tau_m^2)]$$

(iii)  $\tau_m \rightarrow 0$

$$\tilde{X}'_1 \simeq 1/D \quad \tilde{X}''_1 \simeq (1/D)\omega\tau_m.$$

As discussed above, for the PdMn sample the third situation is valid. But for ferromagnetic PdFe the second situation seems to be valid, since the experimental data around and below  $T_c$  shown in figures 1 and 2 indicate the predicted frequency dependence in situation (ii).

The other important conclusion we arrive at is that the third harmonic susceptibility results on these samples support our theoretical conjecture that the non-linear component of the susceptibility diverges while approaching  $T_c$  from above but somehow because of the demagnetizing effect at  $T_c$  it is suppressed and becomes almost zero. Hence a peak is formed just below  $T_c$ . On the other hand, below  $T_c$  dynamical effects prevail. We can prove this by applying higher AC field amplitudes, since at high enough AC field amplitude these dynamical effects can be completely suppressed. For the PdMn sample this observation has been made (see section 3.2).

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