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Critica1 resistivity near the magnetic phase transition in the Heusler compounds Pd_2MnZ (Z = Sn, In, Al)

P. Pureur, G. L. Fraga*, J.V. Kunzler, M. Engel da Silva, R. Menegotto Costa, W.H. Schreiner, D.E. Brandão and M. I. da Costa Jr.
Instituto de Física, Universidade Federal do Rio Grande do Sul, P.O. Box 15051, Porto Alegre, 91500, RS, Brasil

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Abstract We report on the resistivity and the temperature derivative of the resistivity of several Pd-based Heusler compounds near the magnetic phase transition. Ferromagnetic and antiferromagnetic samples are investigated. Several unexpected features are observed in $d\rho/dT$ of these simply idealized magnets. In most cases, a logarithmic singularity could be identified in the paramagnetic fiuctuating regime. In antiferromagnetic Pd_2MnIn, effects associated to disorder and frustration are manifest. Disorder induced features are also affarent in $d\rho/dT$ results below T_C in ferromagnetic Pd_2MnSn.

1. Introduction

Heusler intermetallic compounds have been extensively studied for a long time, and the main concern focussed on the structural[1], magnetic[1], hyperfine field[2] and transport properties[3]. These ternary compounds normally order in the cubic $L2_1$ superstructure shown in fig. 1, and present a magnetic ordered state in the majority of cases. Among the vast number of known Heusler compounds, those of the so-called Pd-family (Pd_2MnZ; Z being an s-p element) are especially interesting. There, the Mn atoms are the only magnetic ones and, through substitution of the Z element, ferromagnetic (Sb, Sn) or antiferromagnetic (In, Al) ground states

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* Present address: Depto. de Física, Universidade Federal de Santa Catarina, 88049 Florianópolis, SC, Brasil.

may be stabilized[1]. Moreover, as the Mn atoms are far apart and are expected to possess virtually zero orbital moment[2], such compounds are often considered as representative of "ideal" local spin magnets[4]. These properties render this series of compounds as interesting experimental model systems for critical behavior studies at the magnetic phase transition. In spite of this, except for an exploratory report[5], no systematic effort devoted to the determination of their critica1 indices is presently available. In this communication, we report on detailed resistivity measurements close to the critica1 temperature in several samples of Pd_2MnZ (Z = Sb, Sn, In, Al) compounds. The measurements are accurate enough to numerically determine the temperature derivative of the resistivity, which gives direct information on the a-critica1 point exponent of the specific heat[6].

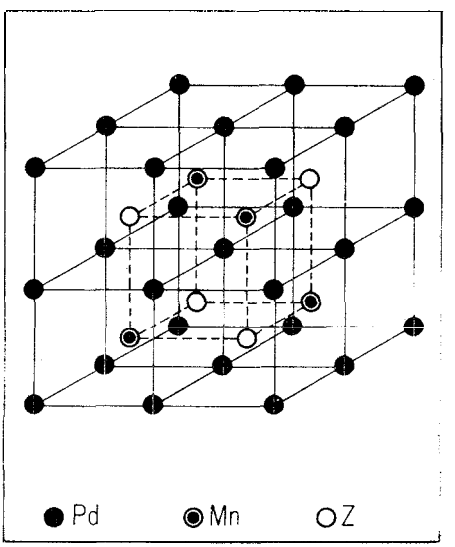

Fig. 1 – The Heusler $L2_1$ structure of Pd_2MnZ.

2. Experimental

Several samples of Pd_2MnZ were prepared following the prescriptions given in ref. 7. Sample #1 of Pd_2MnSn was gently loaned by Dr. C.M. Hurd and its

properties are described in ref. 3. The measured samples are listed in table 1 together with some of their properties.

The resistivity experiments were performed in a temperature interval encompassing T_C by approximately ± 20 K. A four-point AC-lock-in technique was employed with a relative accuracy of about 5 ppm. Temperatures were measured with a Pt sensor to within 1 mK. Measurements were performed while the ternperature drifted at a rate of 8 K/hour or smaller. The experimental points were spaced by temperature intervals ranging from 30 mK near T_C to 300 mK far away from T_C. This allowed us to determine, by a numerical procedure, the temperature derivative of the resistivity, $d\rho/dT$.

Table 1 - Samples investigated in this work. Residual resistivities, critica1 temperatures and structures are also listed. Compounds with Z = Sb, Sn are ferromagnetic while those having Z = In, Al are antiferromagnetic.

Sample	$\rho_0(\mu\Omega.\text{cm})$	T_C(K)	Structure
Pd_2MnSb	6.65	246	$L2_1$
$Pd_2MnSn\#1$	1.17	189	$L2_1$
$PdMnSn\#2$	9.52	182	$L2_1$
$Pd_2MnIn\#1$	30.26	144	$L2_1$ *
$Pd_2MnIn\#2$	34.70	71	B2
Pd_2MnAl	32.2	241	B2

*This sample contains a mall amount of B2 disorder, see section 3.2.1.

3. Results and Discussion

It is theoretically predicted[6,8] and experimentally verified[g] that $d\rho/dT$ near T_C has the same critical singularity as the magnetic specific heat. Therefore, analysis of $d\rho/dT$ results are performed with the usual power law expressions[10] :

$$\frac{d\rho}{dT} = \frac{A^+}{\alpha}(\epsilon^{-\alpha} - 1) + B^+ \quad (T > T_C) \tag{1.a}$$

$$\frac{d\rho}{dT} = \frac{A^-}{\alpha'}(|\epsilon|^{-\alpha'} - 1) + B^- \quad (T < T_C) \tag{1.a}$$

where $\epsilon = (T - T_C)/T_C$, a, a' are the critica1 exponents and A+, $A^- B^+, B^-$ are constants. Scaling theory predicts that $a = a'$ [10]. One notes that, when $a = 0$ eq. (1) is equivalent to a logarithrnic divergence. Performing a non-linear least square fit of the experimental data to eqs. (1), we are able to determine confident values for a, a'. When the data are accurate enough to calculate the second derivative,

$$-\frac{d^2\rho}{dT^2} = \frac{A}{T_C}\epsilon^{-(\alpha+1)} \tag{2}$$

this is concomitantly used an additional support for the determination of the critica1 indices. These may be extracted from logarithmic plots of eq. (2) since good estimates for T_C are possible.

3.1. Ferromagnetic Compounds

3.1.1. Pd_2MnSb

Figure 2 displays the resistivity and $d\rho/dT$ near T_C for a Pd_2MnSb sample. The derivative shows the typical specific heat-like singularity often observed in ferromagnetic transitions. Figure 3.a shows second derivative results. As a first approximation we choose T_C as the temperature of the sharp minimum in $d^2\rho/dT^2$. This gives $T_C = 246.0$ K. Retaining this value for T_C, we perform logarithmic plots of eq. (2) for data in the pararnagnetic range. Results are shown in fig. 3.b where the slope between $0.003 < \epsilon < 0.025$ gives $a = 0.00 \pm 0.03$. This indicates a logarithmic singularity. For smaller ϵ, unavoidable rounding effects due to finite domain size effect and statistical fluctuations of T_C are observed. Fittings of $d\rho/dT$ results to eqs. (1) were perfornied for the $\epsilon > 0$ and $\epsilon < 0$ ranges. Parameters were allowed to vary in wide ranges. The only constraint introduced was the equality of T_C resulting from fits at both sides of T_C. Relevant data are collected in table 2. The logarithmic divergence is confirmed in para- and ferromagnetic fluctuating regimes and the best adjusted T_C is surprisingly close to the value experimentally obtained from the $d^2\rho/dT^2$ results.

Table 2 – Relevant parameters for the samples studied. T_C^{exp} is determined from the minimum in dp^2/dT^2, as in hg. 2.a. T_C^{fit} and a are determined from fits to eqs. (1). E refers to the reduced temperature ranges for fits.

Sample	T_C^{exp} (K)	T_C^{fit} (K)	α	ϵ $(T > T_C)$	$\|\epsilon\|$ $(T < T_C)$
Pd_2MnSb	246.0	246.03 (±0.03)	0.00 (±0.01)	0.003-0.025	-
	-	246.0 (±0.2)	0.00 (±0.01)	-	0.008-0.037
$Pd_2MnSn\#1$	188.6	188.63 (±0.03)	0.00 (±0.02)	0.007-0.055	-
$Pd_2MnSn\#2$	-	181.41 (±0.1)	0.00 (±0.05)	0.033-0.070	-
$Pd_2MnIn\#1$	-	144.0 (±0.2)	-1.3 (±0.1)	-	0.003-0.11
	-	144.0	-1.2 (±0.2)	0.019-0.17	-
$Pd_2MnIn\#2$	70.6	-	-	-	
Pd_2MnAl	240.8	240.76 (±0.03)	0.00 (±0.01)	0.008-0.041	

3.1.2. Pd_2MnSn

Resistivity and $d\rho/dT$ results for samples #1 and #2 are shown in figures 4.a and 4.b, respectively. The main difference of these samples is the amount of crystalline disorder which is manifest in their quite distinct values for the residual resistivity, as listed in table I. In the paramagnetic range, $d\rho/dT$ in both samples may be fitted to $\ln\epsilon$ in a large reduced temperature range (see table 2). Logarithmic plots of the second derivative confirm the value $a = 0$, as shown by the straight line in figure 5, where several results for sample #1 are plotted. These measurements were performed upon application of small magnetic fields, which do not alter the fluctuation resistivity. For sample #1, the experimental criterion of defining T_C as the minimum of $d^2\rho/dT^2$ gives values quite close to T_C obtained

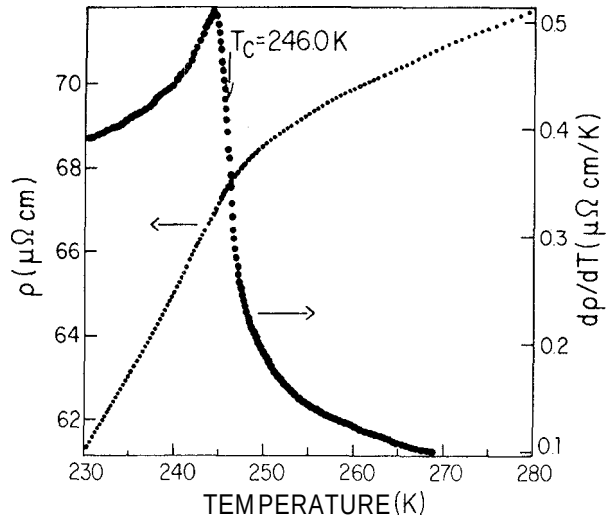

Fig. 2 – Resistivity and $d\rho/dT$ for Pd_2MnSb. For clarity, only part of the experimental data are plotted.

from best fits to eqs. (1). However, for sample #2 the minimum in $d^2\rho/dT^2$ is too much rounded for a precisely indicating a typical temperature.

Below the critica1 temperature, an unusual minimum appears in $d\rho/dT$ for both samples, as shown in figs. 4. In the more disordered sample #2 the minimum goes through negative values, traducing the bump observed in the resistivity curve. Because of this complex structure we are unable to fit data in the ferromagnetic range. We note, however, that the minimum feature may be gradually supressed by applying a small magnetic field parallel to the current, until it is completely removed at 20 G for sample #1 and 80 G for sample #2 (see fig. 1 of reference 5). The origin of this complex behavior below T_C is difficuit to establish. One possibilty comes from mean field effects associated with the growth of the spontaneous magnetization[10]. In Heusler compoiinds, however, ferro- and antiferromagnetic ground states are close in energy. Specially in the Pd_2MnSn system, it is known that the saturation magnetization is remarkably dependent upon cold

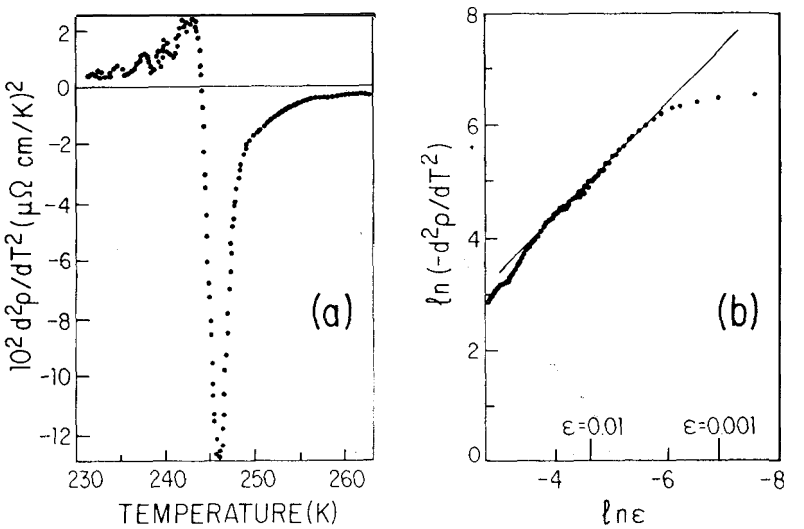

Fig. 3 – Second derivative $d^2\rho/dT^2$ for Pd_2MnSb (a) as measured and (b) in logarithmic plots with respect to the reduced temperature $\epsilon = (T - T_C)/T_C$. In part (b) are shown results of the paramagnetic range and the straight line corresponds to a slope equals to 1, indicating $a = 0$ in eq. (2).

work[11]. This is due to the formation of extended defects like dislocations and antiphase boundaries where the Mn-Mn distance shortens considerably, giving place to antiferromagnetic couplings. Close to T_C, it is possible that antiferromagnetic ordered regions larger than the electron mean free-path nucleate around these defects and persist for fluctuation times larger than the electron relaxation time. Antiferromagnetic ordering often produces a minimum in the resistivity just below T_C because of the opening of magnetic superzone gaps on the Fermi surface. As the minima shown in $d\rho/dT$ and ρ (for sample #2) are clearly related to disorder, we are inclined to believe that this is the best description of the complex resistivity behavior observed close to T_C in our Pd_2MnSn samples.

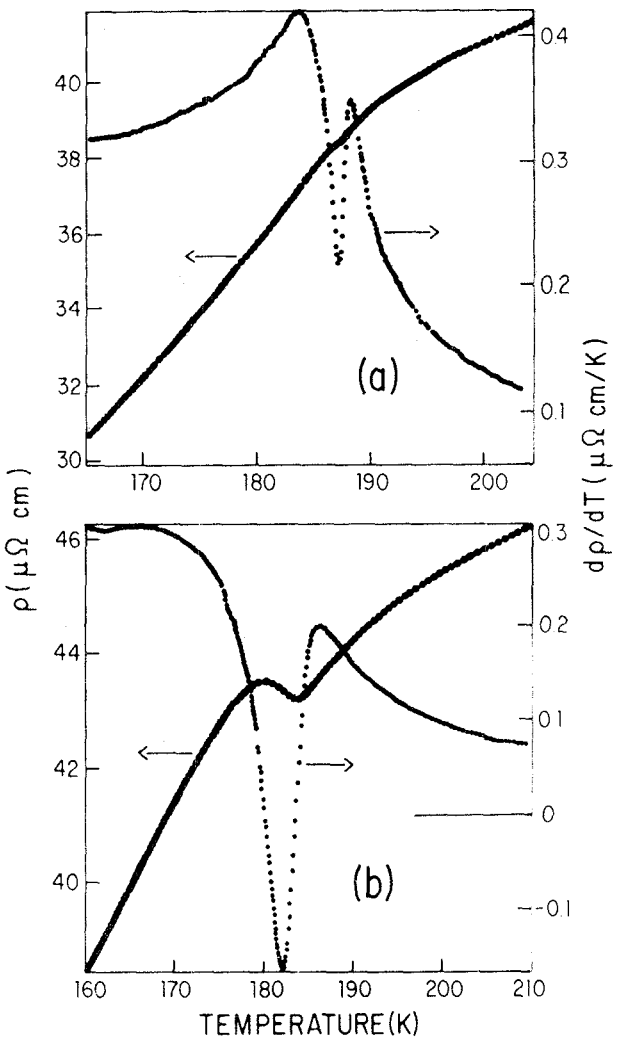

Fig. 4 – Resistivity and $d\rho/dT$ for (a) sample #1 and (b) sample #2 of Pd_2MnSn. For clarity, part of the resistivity points is not plotted.

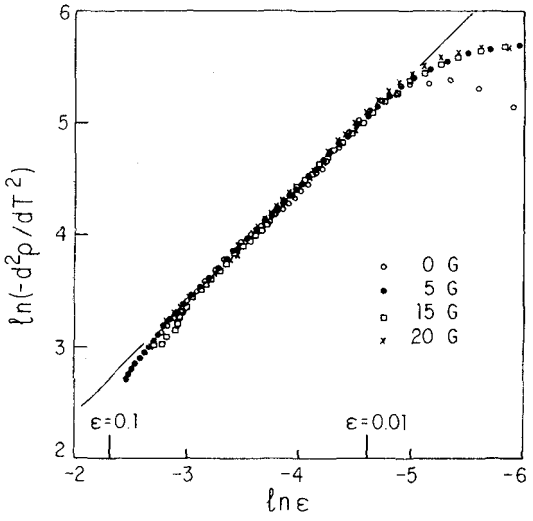

Fig. 5 - Logarithmic plots of the second derivative with respect to the reduced temperature in the paramagnetic regime of Pd_2MnSn#1. Measurements are performed in the magnetic fields quoted. The straight line corresponds to a $\pi/4$ inclination.

3.2. Antiferromagnetic Compounds

3.2.1. Pd_2MnIn

In these compounds a very special kind of crystalline disorder may occur, namely the site interchange of Mn and In atoms. When the site occupancies of the Mn and Z elements are totally random, the modified structure is called **B2.** This kind of disorder often depends on the thermal history of the sample and normally affects the magnetic properties of the Heusler compounds severely, because of its tendency to shorten Mn-Mn distances. We investigated two Pd_2MnIn samples. One of them orders in the $L2_1$ structure with some minor amount of B2-like disorder (sample #1). The second is completely B2 (sample #2). Figure 6 shows the results for both samples. In the $L2_1$ sample we obtain an invertedly by peaked for $d\rho/dT$, as expected on theoretical grounds[12]. However, the exponents found in fits to eqs. (1) are very large ($\alpha < -1$) both above and below T_C (see table 2).

Similar values for a were extracted from $d\rho/dT$ results close to the Curie temperature of the reentrant systems a-$\underline{Fe}Zr$ and $\underline{Ni}-Mn$[13]. There, the large values of the a-exponents were supposed to be related to the disordered and frustrated magnetic state. The same may he the case in Pd_2MnIn compounds. Indeed, when the amount of B2 disorder is increased, as shown in fig. 6.b, the Néel temperature is lowered and $d\rho/dT$ develops a rounded positive-peaked picture which qualitatively reminds the magnetic specific heat of the spin glasses[14], except for the sharp anomaly denoting the transition.

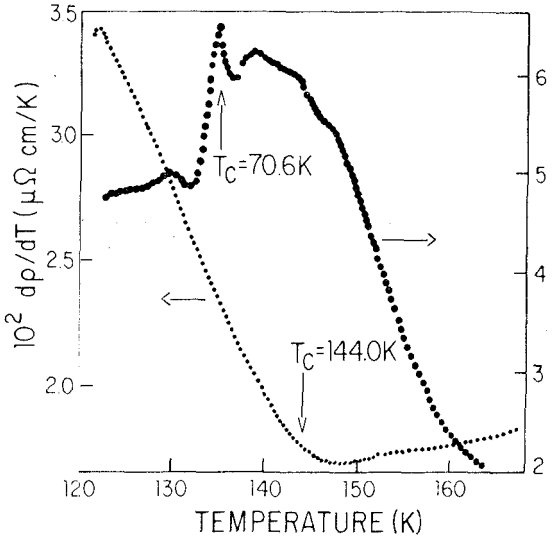

Fig. 6 - Derivative results for sampie #1 (small dots) and sample #2 (full circles) of Pd_2MnIn.

3.2.2. Pd_2MnAl

This compound orders only in the B2 structure. Its $d\rho/dT$ singularity at the Néel temperature is positive-peaked as for the ferromagnetic samples. In the paramagnetic range we could identify a $\ln\epsilon$ regime extending from $\epsilon = 0.008$ to

$\epsilon = 0.04$. For $T < T_C$, $d\rho/dT$ is almost flat and the temperature dependence of $d\rho/dT$ is difficult to determine.

4. Conclusions

Although the Heusler compounds of the Pd_2MnZ series are generally thought as being representative of ideal local moment magnets, our $d\rho/dT$ results near T_C reveal quite unexpected characteristics. One is the rather extended logarithmic temperature dependence of $d\rho/dT$ in the paramagnetic regime for most of the investigated systems. On the basis of renormalization group calculations[15] one would expect that for a Heisenberg ferromagnetic $\alpha \cong -0.11$. From a theoretical point of view, $\alpha = 0$ would be expected for XY spins in 3D systems or Ising spins in 2D systems[15,16]. However, it is hard to model Heusler compounds in such low dimensionâl situations. A possibility might be to argue that extended planar defects as antiphase boundaries and dislocations, which are characteristic of these compounds[17], are the geometrical entities governing the critical energy fluctuations. We note that logarithmic singularities have already been observed in $d\rho/dT$ measurements in other localized spin magnets[18].

In the antiferromagnetic Pd_2MnIn samples, fluctuation behavior seems to be dependent of disorder and frustration. Disorder also affects strongly the results below T_C in the Pd_2MnSn samples, where a complex behavior in $d\rho/dT$ is noticeable.

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Resumo

Nós apresentamos resultados de resistividade elétrica, e sua derivada com relação a temperatura, de vários compostos de Heusler da família do Pd, em temperaturas próximas à transição de fase magnética. Sistemas ferromagnéticos e antiferromagnéticos foram investigados. Várias características inesperadas foram observadas em $d\rho/dT$ destas amostras, as quais são geralmente descritas como magnetos idealmente simples. Na maioria dos casos, identificamos uma singularidade logaritmica no regime de Autuações da fase paramagnética. No antiferromagneto Pd_2MnIn aparecem claramente efeitos associados com desordem e frustração.

Efeitos induzidos por desordem também são visíveis nos resultados de $d\rho/dT$ no ferromagneto Pd_2MnSn.