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LETTER TO THE EDITOR

Thermal irreversibility effects close to the antiferromagnetic transition temperature in $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems

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Abstract. The electrical resistivity of $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems has been measured between 1.7 and 300 K. We observe, for the first time in Pr-based ternary systems, thermal hysteresis effects, at temperatures below their antiferromagnetic ordering and temperatures, in a narrow concentration range of $0.075 \leqslant x \leqslant 0.15$. This hysteresis effect is also corroborated by the magnetization results. It could be related either to spin reorientation effects or to the quadrupolar strains distorting the lattice.

The low-temperature magnetic behaviour of Pr-based compounds can arise from two sources. The first source is the usual magnetic interaction between the 4f and the conduction electrons as seen also in several well studied Ce-based Kondo lattice (KL) systems. This magnetic interaction often leads to long-range magnetic ordering effects in Pr-based compounds. Though the basic magnetic interactions in Pr systems is intrinsically similar to the Ce-based KL ones, the differences in the Pr 4f radial wavefunction extension with respect to Ce and and the specific crystal field splitting of the Pr energy levels give rise to novel features in their magnetic behaviour. For example, the occurrence of the Kondo effect is quite rare in Pr compounds. Furthermore, the magnetism of Pr systems is more itinerant than localized compared with the Ce ones. The second source is the quadrupolar interaction, which is purely non-magnetic in nature. This interaction can lead to a preferential alignment of non-spherical 4f shells of the Pr atoms. This alignment could cause a lattice strain and lead to structural transformation effects [1]. It could also induce long-range magnetic ordering effects because the quadrupole and the magnetic moments are coupled to each other. The study [2–4] of Pr systems has therefore been of interest in recent years.

With this in mind, we have studied the pseudoternary systems $Pr(Cu_{1-x}Ni_x)_2Si_2$. Both the parent compounds $PrCu_2Si_2$ ($x = 0$) and $PrNi_2Si_2$ ($x = 1$) exhibit antiferromagnetic (AF) ordering [5, 6] at temperatures $T_N = 21$ and 18 K, respectively. However, their magnetic structures are quite different. In both these compounds, the ordering of the Pr moments, within the a–b basal plane of the tetragonal crystal structure, is ferromagnetic. The moments are directed along the c axis. For $PrCu_2Si_2$, these ferromagnetic planes are coupled to each other antiferromagnetically with the sequence $+ - +-$ along the c axis. Also, the AF ordering is commensurate (C) with the lattice periodicity. However, for $PrNi_2Si_2$ ($T < T_N$), the moments modulate sinusoidally, along the c axis, with a period which is incommensurate (IC) with the lattice periodicity. Furthermore, the values of T_N for both these compounds are even larger than those for their isostructural Gd compounds in violation [5, 6] of the deGenne's scaling law.

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All the alloys were prepared in an arc furnace under flowing argon conditions. The alloys have been annealed at 800 °C for one week, similar to that carried out in an earlier study [7]. They were found to crystallize in the $ThCr_2Si_2$ type tetragonal structure. The lattice spacings (table 1) a and c were both found to decrease with x, thus decreasing the unit cell volume.

Table 1. Lattice parameters a and c and the unit cell volume V for the $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems.

System $Pr(Cu_{1-x}Ni_x)_2Si_2$	a (Å)	c (Å)	V (Å^3)
$x = 0$	4.0856±0.0009	9.9292±0.0038	165.7±0.1
0.04	4.0851±0.0017	9.9331±0.0050	165.7±0.1
0.075	4.0845±0.0026	9.9245±0.0094	165.6±0.1
0.15	4.0817±0.0014	9.8749±0.0052	164.5±0.1
0.25	4.0787±0.0009	9.8173±0.0047	163.3±0.1
0.40	4.0751±0.0013	9.7695±0.0047	162.2±0.1
0.75	4.0713±0.0008	9.6733±0.0028	160.3±0.1
0.90	4.0632±0.0007	9.6425±0.0023	159.2±0.1
1.0	4.0469±0.0008	9.6212±0.0025	157.6±0.1

As the temperature is decreased, the ρ data (figure 1) for the $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems show (figure 2) a minimum at T^{ρ}_{min} (table 2), followed by a maximum at T^{ρ}_{max} (table 2). The value of T^{ρ}_{min} is close (table 2) to T_N obtained [3, 6, 7] from neutron and susceptibility measurements. The most interesting observation, from our studies, is the occurrence of thermal hysteresis effects in the ρ curve, occurring near T^{ρ}_{max}, for the $x = 0.075$ and $x = 0.150$ systems only. The magnetization M data (obtained using a SQUID magnetometer—Quantum Designs, USA), also corroborates the ρ findings by exhibiting (figure 3) similar thermal hysteresis effects, below T_N, for the $x = 0.075$ system, but not for the $x = 0.04$ system. The ρ and M behaviours thus show that the thermal hysteresis effects occur only within a narrow interval of x (shaded portion—figure 4) where the value of T_N is found to decrease rapidly with increase in x. This hysteresis behaviour has not been reported so far, even though similar ρ curves have been reported in an earlier study [7].

Table 2. Characteristic temperatures for the $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems, as obtained from our ρ studies.

System $Pr(Cu_{1-x}Ni_x)_2Si_2$	T^{ρ}_{min} (K)	T^{ρ}_{max} (K)	T_N (K)
$x = 0$	21	19	21[a]
0.04	16	12	—
0.075	12	7	13[b]
0.15	9	5	10[b]
0.25	6	3	9[b]
0.4	9	5	—
0.75	14	10	16[b]
0.9	15	10	—
1.0	17	15	18[c]

[a] [6].
[b] [7].
[c] [3].

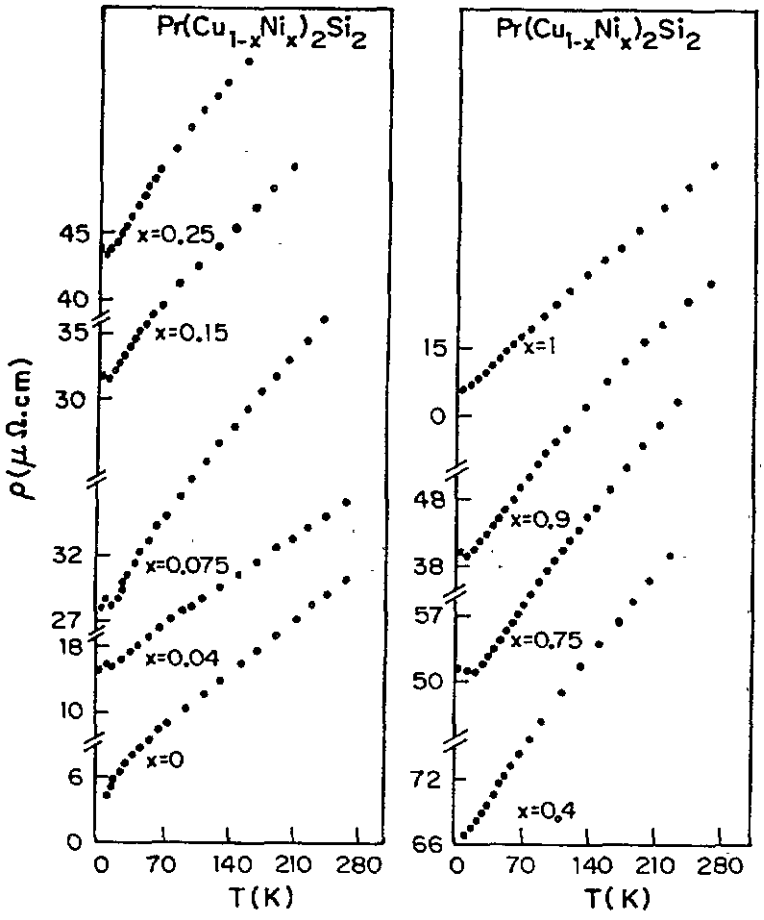

Figure 1. ρ versus T curves from 4.2 to 300 K for the $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems.

This effect could arise from the following factors. For the $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems, the C and the IC antiferromagnetic behaviour of the end compounds ($x = 0$, $x = 1$), respectively, could become energetically close to each other. As a result, for the intermediate values of x ($0.075 \leqslant x \leqslant 0.15$), for $T \leqslant T_N$, the C structure could begin to develop first. As the temperature is gradually decreased below T_N, the value of ρ increases, possibly due to opening of the AF gap [7], and passes through a maximum (figure 2) at T_R. The subsequent decrease in ρ, below T_R, could be related to the gradual conversion of the C spin structure phase to an IC one. For our systems ($0.075 \leqslant x \leqslant 0.15$), within a short temperature interval ($\simeq$ 1–2 K) around T_R, the IC phase would become fully formed. The development of this IC phase within the C phase, we believe, is associated with the thermal hysteresis (figure 2). Whereas, in contrast to the above, the $x < 0.075$ and the $x > 0.15$ systems probably develop either the C or the IC phase, respectively. Neutron studies would be required to confirm these viewpoints.

The changes in the magnetic structure (for $0.075 \leqslant x \leqslant 0.15$), below T_N, are referred to as spin-reorientation effects and have also been observed in several systems like $PrGa_2$, Pr_3Al and $TbNi_2Si_2$ also. In $PrGa_2$, a long-period sinusoidally modulated AF structure (for $T_R < T < T_N$) changes [2] to a C–AF structure below T_R. In Pr_3Al, an AF phase between

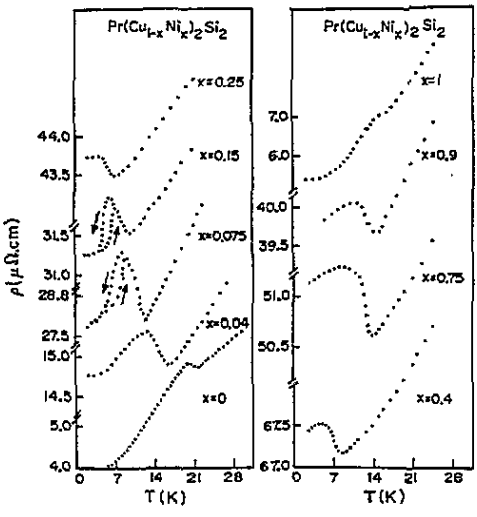

Figure 2. Expanded plots for the ρ versus T curves for the $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems. Note the hysteresis effects in the $x = 0.075$ and $x = 0.15$ systems.

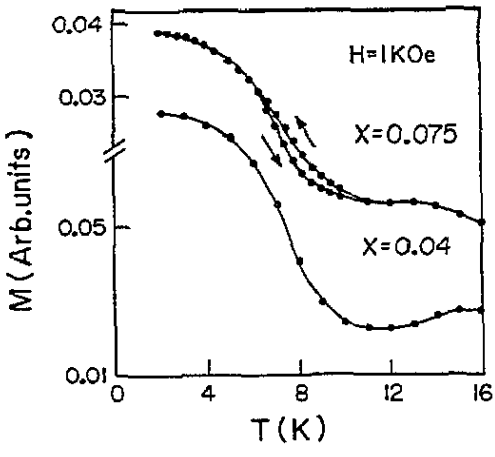

Figure 3. M versus T curves for the $x = 0.04$ and the $x = 0.075$ systems. Note the thermal hysteresis effects in $x = 0.75$ system which is absent in the case of $x = 0.04$ system. The solid lines are drawn to guide the eye.

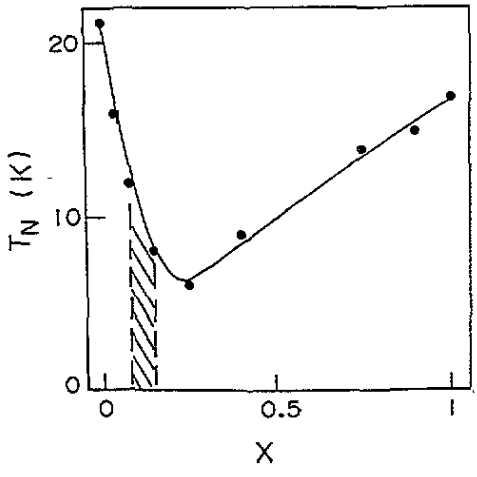

Figure 4. T_N versus x curve for $Pr(Cu_{1-x}Ni_x)_2Si_2$ systems. The shaded portion denotes the range of x in which the thermal hysteresis is observed. The solid line is drawn to guide the eye.

$T_R < T < T_N$ changes [8] to a ferrimagnetic state below T_R, whereas in $TbNi_2Si_2$, an IC structure between $T_R < T < T_N$ changes [9] to a C structure below T_R. Another common feature, in these compounds, is the presence of thermal hysteresis effects [2, 8, 9] around T_R. We note that this hysteresis effect could also arise from lattice distortions, due to the quadrupolar alignment of the Pr 4f shells. Such quadrupole induced hysteresis effects have also been observed [10–12] in $PrCu_2$ and $PrPb_3$.

In conclusion, thermal hysteresis effects have been observed for the first time in

$Pr(Cu_{1-x}Ni_x)_2Si_2$ systems within a narrow range of $0.075 \leqslant x \leqslant 0.15$. These may be related with either spin-reorientation or quadrupolar induced effects.

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