

Magnetism and charge density waves in RNiC₂ (R = Ce, Pr, Nd)

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We have compared the magnetic, transport, galvanomagnetic and specific heat properties of CeNiC₂, PrNiC₂ and NdNiC₂ to study the interplay between charge density waves and magnetism in these compounds. The negative magnetoresistance in NdNiC₂ is discussed in terms of the partial destruction of charge density waves and an irreversible phase transition stabilized by the field induced ferromagnetic transformation is reported. For PrNiC₂ we demonstrate that the magnetic field initially weakens the CDW state, due to the Zeeman splitting of conduction bands. However, the Fermi surface nesting is enhanced at a temperature related to the magnetic anomaly.

I. INTRODUCTION

The interaction between charge density waves (CDW) and different types of orderings such as superconductivity¹⁻³, spin density waves⁴⁻⁶ and magnetism⁷ has been a long standing area of interest. Magnetic order or applied magnetic field have been found to impact the CDW state through changing the geometry of the Fermi surface (FS). The effect can be destructive due to the disturbance of the FS nesting caused by the magnetic field-induced splitting of the conduction bands or modification of the electronic structure due to a magnetic transition⁸. Alternatively, a constructive effect has been observed in a group of materials, in which this FS transformation leads to the enhancement of the nesting conditions or when the nesting vector has the ability to adapt to the evolution of the Fermi surface⁹⁻¹⁵. Recently, much attention of the researchers exploring the coupling between CDW, superconductivity and magnetic order has been devoted to the two families of ternary compounds: M₅Ir₄Si₁₀, (where M = Y, Dy, Ho, Er, Tm, Yb or Lu)¹⁶⁻²⁴ and RNiC₂, (where R = La, Ce, Pr, Nd, Sm, Gd or Tb)^{25,26}. Most of the members of the latter family exhibit the Peierls transitions towards the charge density wave state²⁷. The relevance of a Peierls instability has been confirmed for R = Gd, Tb, Nd, Pr and Sm, while the LaNiC₂ and CeNiC₂ compounds do not show any anomalies that could be attributed to CDW²⁸⁻³². Instead, LaNiC₂ is an unconventional noncentrosymmetric superconductor with $T_c = 2.7$ K³³⁻³⁵. Next to the CDW, the members of the RNiC₂ family show a wide range of magnetic orderings originating from the RKKY interaction between local magnetic moments and conduction electrons^{36,37}. The ground state of RNiC₂ depends on the rare-earth atom marked in the above formula by R: CeNiC₂, NdNiC₂, GdNiC₂ and TbNiC₂ show the antiferromagnetic character^{34,38-42}, SmNiC₂ is a ferromagnet, while the PrNiC₂ compound has been identified as a van Vleck paramagnet⁴³. This rich variety of the types of magnetic ordering shown by the RNiC₂ family members motivated us to explore the interplay of charge density waves and various magnetic ground states. Here, we compare the physical properties of three isostructural, yet highly dissimilar compounds:

NdNiC₂, PrNiC₂ and CeNiC₂. The first compound, NdNiC₂ shows the Peierls instability with $T_P = 121$ K and antiferromagnetic ordering with $T_N = 17$ K. The second, PrNiC₂ undergoes the CDW transition at $T_P = 89$ K and instead of long range magnetic ordering, shows a magnetic anomaly at $T^* = 8$ K. The last compound, CeNiC₂ becomes an antiferromagnet at $T_N = 20$ K and does not exhibit the CDW transition.

II. EXPERIMENTAL DETAILS

The polycrystalline samples of RNiC₂ (where R = Ce, Pr, and Nd) were synthesized by arc-melting the stoichiometric amounts of pure elements: Ni (4N), C (5N) and Ce (3N), Pr (3N), Nd (3N) in a high purity argon atmosphere. Small excess of Ce, Pr, Nd ($\approx 2\%$) and C ($\approx 5\%$) was used to compensate the loss during arc-melting. To obtain good homogeneity of samples, the specimens were turned over and remelted four times in a water-cooled copper hearth. A zirconium button was used as an oxygen getter. The buttons obtained from the arc-melting process were wrapped in tantalum foil, placed in evacuated quartz tubes, annealed at 900°C for 12 days and cooled down to the room temperature by quenching in cold water. Overall mass loss after the melting and annealing processes were negligible ($\approx 1\%$).

The low temperature experiments were performed with a Quantum Design physical properties measurements system (PPMS) allowing for the application of a magnetic field as large as 9 T. Thin Pt wires ($\phi = 37$ μm) serving as electrical contacts for transport and Hall measurements were spark-welded to the polished sample surface. A standard four-probe contact configuration was used to measure resistivity. A magnetic field was applied perpendicularly to the current direction. The Hall voltage was collected in reversal directions of magnetic field in order to remove the parasitic longitudinal magnetoresistance voltage due to misalignment of electrical contacts. The specific heat measurements were performed using the dual slope method on flat polished samples. Magnetization measurements were carried out using the ACMS susceptometry option of the PPMS system. Pieces of the samples were fixed in standard polyethylene straw holders.

III. RESULTS AND DISCUSSION

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91 The phase composition and crystallographic structure
 92 of the samples were checked by powder X-ray diffraction
 93 (pXRD) at room temperature. The pXRD analysis
 94 shows that all observed peaks for NdNiC₂ and PrNiC₂ are
 95 successfully indexed in the orthorhombic CeNiC₂-type
 96 structure⁴² with a space group Amm2 (# 38), which con-
 97 firms the phase purity of the obtained samples. Only for
 98 the CeNiC₂ sample, additional reflections corresponding
 99 to a small amount of the secondary phase⁴⁴ CeC₂ are ob-
 100 served. The lattice parameters were determined from the
 101 LeBail profile refinements of the pXRD patterns carried
 102 out using FULLPROF software⁴⁵. The obtained values
 103 of the lattice constants, shown in Table I are in good
 104 agreement with those reported in the literature^{39,43,46,47}.

105

TABLE I. Lattice constants, unit cell volume and the parameters of the LeBail refinements for CeNiC₂, PrNiC₂ and NdNiC₂, at room temperature.

	CeNiC ₂	PrNiC ₂	NdNiC ₂
a (Å)	3.8753(2)	3.8239(5)	3.7834(1)
b (Å)	4.5477(2)	4.5428(8)	4.5361(1)
c (Å)	6.1601(3)	6.1448(1)	6.1285(1)
V (Å ³)	108.565(8)	106.746(3)	105.178(3)
R _p	12.3	7.51	8.35
R _{wp}	16.5	10.1	10.8
R _{exp}	11.49	7.54	7.7
χ ²	2.05	1.81	1.96

106

107

108 The temperature dependence of the magnetic suscep-
 109 tibility (χ) measured at 1 T applied magnetic field is
 110 presented in Figure 1. All three compounds show para-
 111 magnetic behavior at high temperatures. The χ(T) data
 112 were fitted using the modified Curie-Weiss expression:

$$\chi(T) = \frac{C}{T - \Theta_{CW}} + \chi_0 \quad (1)$$

113 where C is the Curie constant, Θ_{CW} is the Curie-Weiss
 114 temperature, and χ₀ is the temperature-independent sus-
 115 ceptibility resulting from both sample (Pauli and Van
 116 Vleck paramagnetism, Landau diamagnetism) and sam-
 117 ple holder (small diamagnetic contribution of sample
 118 straw assembly). Having estimated the C parameter and
 119 assuming that the magnetic moment originates from R³⁺
 120 ions only, one can calculate the effective magnetic mo-
 121 ment using the relation shown in Equation 2:

$$\mu_{eff} = \sqrt{\frac{3Ck_B}{\mu_B^2 N_A}} \quad (2)$$

122 where k_B is the Boltzmann constant, μ_B is the Bohr
 123 magneton, and N_A is Avogadro's number. The result-
 124 ing effective magnetic moments of CeNiC₂, PrNiC₂ and

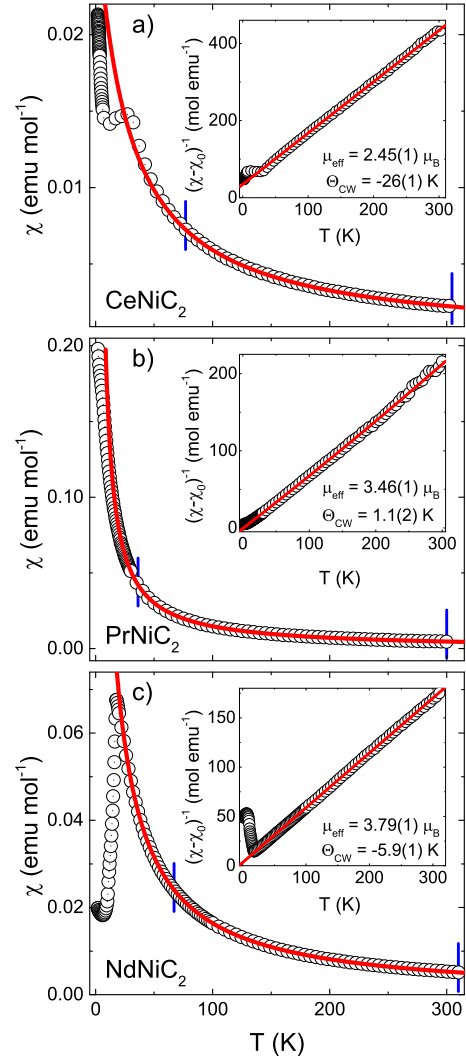


FIG. 1. Magnetic susceptibility of CeNiC₂ (a), PrNiC₂ (b), and NdNiC₂ (c) at applied magnetic field μ₀H = 1 T (open circles). Red lines show fits using the modified Curie-Weiss expression (Eq. 1). Insets show inverse susceptibilities displaying linear temperature dependence in agreement with the Curie-Weiss law (Eq. 1). Blue ticks mark the used fitting ranges. The effective magnetic moments extracted from fits agree with the values expected for free trivalent R ions. Low-temperature part of susceptibility for PrNiC₂ is presented in Fig. 2

125 NdNiC₂ are consistent with the values expected for free
 126 R³⁺ ions⁴⁸. The negative sign of Θ_{CW} obtained for the
 127 Ce- and Nd-bearing compounds (-26 K and -5.9 K, re-
 128 spectively) indicate an effectively antiferromagnetic cou-
 129 pling between the magnetic moments. In the case of
 130 PrNiC₂, the absolute value of Θ_{CW} is close to 0 sug-
 131 gesting the weakness or absence of magnetic interactions
 132 down to 2 K.

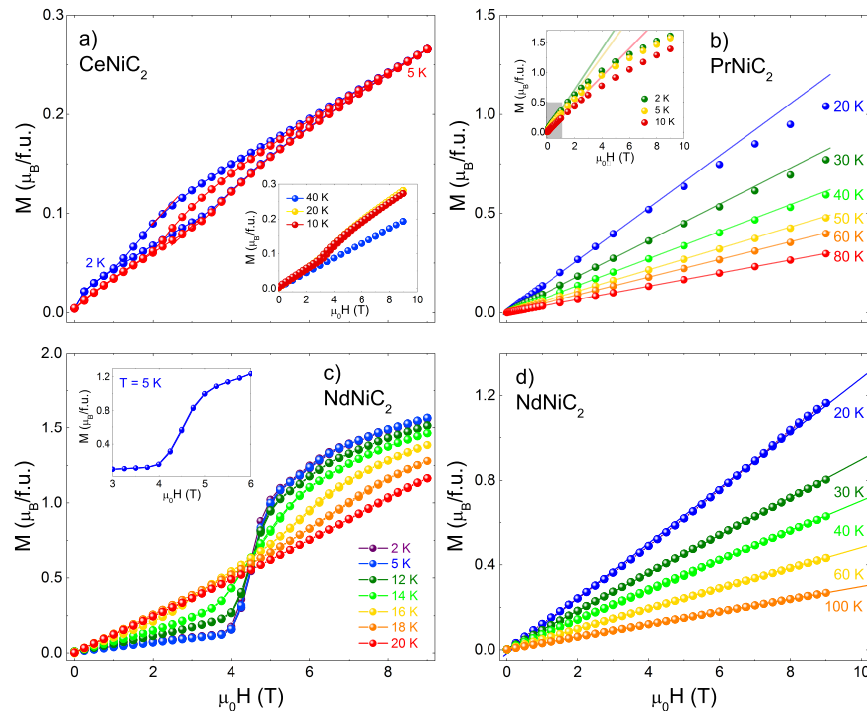


FIG. 2. Panel a) Magnetization vs. applied magnetic field ($M(H)$) measured for CeNiC_2 at 2 and 5 K (below the Néel temperature $T_N = 19$ K) showing a hysteretic behavior probably due to a field-induced magnetic transition. The inset presents the magnetization at 10, 20, and 40 K. While the magnetization at $T \geq 40$ K (above the AFM transition) is a linear function of applied field, in the vicinity (20 K) and below the T_N an upturn is seen around 3 T, suggesting the field-induced magnetic transition suppressing the AFM order. Panel b) presents $M(H)$ curves for PrNiC_2 showing linear character down to 40 K. Below that temperature the curves start to saturate in high magnetic fields. At the lowest temperatures (2, 5, and 10 K; see the inset) the deviation from linearity is clear above 1-2 T. Straight lines are least-squares linear fits to the low-field (below 1 T) magnetization data. Gray shading in the inset marks the fitting range used. Panel c) shows the low-temperature $M(H)$ data for NdNiC_2 . At 20 K (above the $T_N = 17$ K) the curve is linear up to 9 T while below this temperature an upturn is observed above approx. 4 T. In the temperatures lower than T_N the magnetization below approx. 4 T is visibly suppressed due to AFM ordering of the magnetic moments. At 4 T a magnetic order-order transition results in rapid increase in magnetization. The inset shows magnetization around the field-induced magnetic transition at 5 K showing no sign of hysteresis. Panel d) presents magnetization of NdNiC_2 between 20 and 100 K, showing a linear character up to 9 T. Straight lines are least-squares linear fits to the low field data.

133 It is worth noting that the measured susceptibility of
 134 PrNiC_2 is well reproduced by the modified Curie-Weiss
 135 equation, yielding reasonable values of C , Θ_{CW} , and χ_0
 136 and suggesting that the contribution of Pr^{3+} local mo-
 137 ments is the dominant part of magnetic susceptibility
 138 above 35 K. The Van Vleck paramagnetic contribution
 139 reported by Onodera et al.⁴³ is in our case well modeled
 140 by the temperature-independent term χ_0 .

141 Upon crossing the Néel temperature $T_N = 17$ K, the
 142 magnetic susceptibility of NdNiC_2 drops rapidly. A sim-
 143 ilar drop, yet much less pronounced, is seen also in
 144 CeNiC_2 below $T_N = 19$ K. The susceptibility of PrNiC_2
 145 shows no clear sign of a magnetic transition above 2 K,
 146 in agreement with previous reports^{37,43}, however a small
 147 kink in the curve is seen at $T^* \approx 8$ K (see Fig. 3),
 148 consistent with the decrease in magnetization along the
 149 a crystallographic axis seen at this temperature by On-

150 odera et al.⁴³). The underlying cause for this magnetiza-
 151 tion anomaly is not clear, but may suggest some type of
 152 electronic or crystal structure transition, resulting in the
 153 decrease of Pauli or Van Vleck paramagnetic susceptibil-
 154 ity.

155 Magnetization vs. applied field ($M(H)$) for CeNiC_2 ,
 156 PrNiC_2 , and NdNiC_2 is presented in Figure 2. For
 157 CeNiC_2 (Fig. 2a) the magnetization is linear above T_N ,
 158 with an upturn developing above approx. 4 T in the
 159 lower temperatures. Below the second transition temper-
 160 ature ($T_t = 7$ K) hysteresis is observed in $M(H)$. Even
 161 at 9 T applied magnetic field, the magnetization reaches
 162 only $0.27\mu_B$ which is ca. 13% of the expected satura-
 163 tion magnetization for Ce^{3+} ion $gJ = 2.14\mu_B$ (where
 164 $g = \frac{4}{5}$ is the Lande g -factor, and $J = 4$ is the total an-
 165 gular momentum)⁴⁸. The magnetization at 2 K and 9
 166 T for CeNiC_2 is however approximately half of the ob-

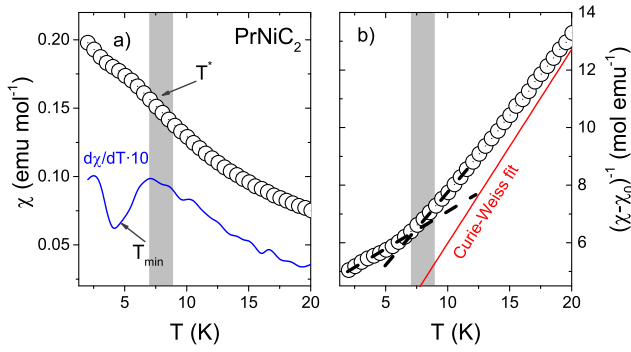


FIG. 3. a) Low-temperature dc magnetic susceptibility of PrNiC₂ measured at 1 T applied field showing a slight upturn around 7 K, below the magnetic anomaly temperature T^* (see text). The differential of the dc susceptibility (blue line) shows a minimum around 4 K. b) Inverse magnetic susceptibility of PrNiC₂ corrected for the temperature independent contributions χ_0 . Red line shows the Curie-Weiss fit from Fig. 1 b). Dashed lines are a guide for the eye.

served saturation moment for a pure Ce metal which is only $0.6\mu_B$ ⁴⁸.

For PrNiC₂, $M(H)$ is roughly linear up to 9 T applied field at temperatures above 40 K (see Fig. 2b), below which the curves start to slightly deviate from linearity. At 10 K and below (Inset of Fig. 2b) the deviation is more pronounced and the curves start to saturate. At 2 K and 9 T applied field the $M(H)$ of PrNiC₂ reach approx. $1.5\mu_B$, which is half of the expected saturation magnetization for Pr³⁺ ion $gJ = 3.20\mu_B$ ⁴⁸.

In case of NdNiC₂, the magnetization curves are linear down to 20 K (Fig. 2c and d). Below the T_N the $(M(H))$ is strongly suppressed, but above 4 T a sudden upturn is observed, resulting from field-induced magnetic order-order transition that reduces the AFM compensation of local moments. Similar transitions have been previously observed in GdNiC₂⁴⁹. Above the transition the $M(H)$ curves start to saturate, reaching $1.6\mu_B$ in 9 T at 2 K, about one half the saturation magnetization for Gd ion ($gJ = 3.27\mu_B$ ⁴⁸). The magnetization loop shows no trace of hysteresis at the AFM-FM transition as it is presented in the inset of Fig. 2c.

The real part of the ac magnetic susceptibility of CeNiC₂ and NdNiC₂ shows a drop at the Néel temperature T_N of 19 and 17 K, respectively (see Fig. 4a,c), in agreement with previous reports⁴³. Below T_N both compounds undergo further magnetic transitions. In CeNiC₂ a sudden drop of susceptibility is seen at $T_t = 7$ K followed by a pronounced upturn. The change in magnetic order below 10 K was previously observed by magnetization, specific heat and NMR measurements^{43,46}. An additional small upturn around 29 K results from the presence of a minor quantity of the antiferromagnetic CeC₂ impurity phase⁴⁴ ($T_N = 30$ K), observed in XRD measurements. In NdNiC₂ a small feature is seen around 4 K

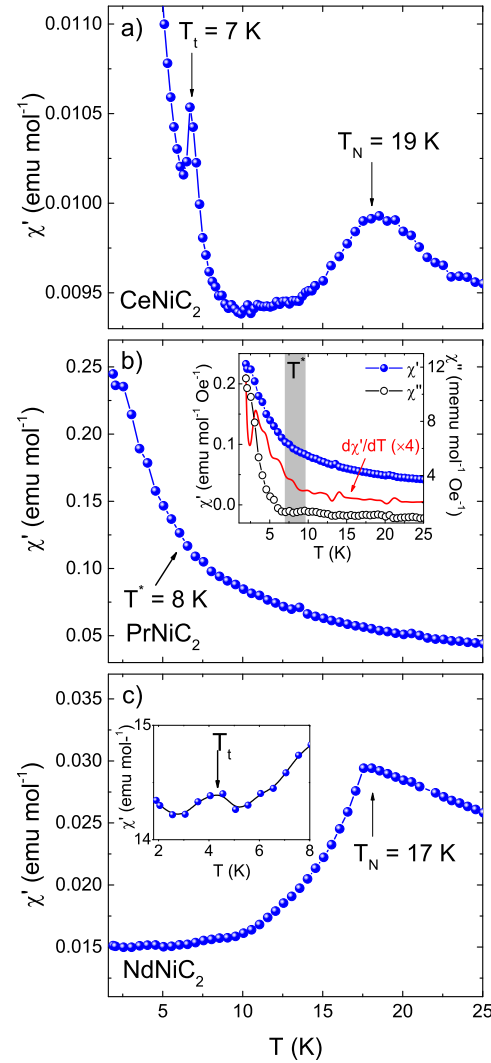


FIG. 4. Real part of ac magnetic susceptibility of a) CeNiC₂, b) PrNiC₂, c) NdNiC₂ measured in a constant field of 5 Oe with 3 Oe, 1 kHz excitations. Blue arrows on panel a indicate the transition to an AFM state at $T_N = 19$ K and order-order transition at approx. 7 K. Inset of panel b presents the comparison of real and imaginary parts of the ac susceptibility (blue and black points, respectively) and the derivative of the real part (red line). The value of derivative is negative and decreases with decreasing temperature. In panel c the $T_N = 17$ K is defined as a position of the drop of susceptibility at the AFM transition. Inset shows a small jump around 4 K that is attributed to magnetic order-order transition.

(see the inset of Fig. 4c) that was reported by Onodera *et al.*⁴³. The ac susceptibility of PrNiC₂ shows no clear sign of magnetic transition, however the slightly saturating dependency of χ' and its derivative $d\chi'/dT$ resembles the results obtained for the Pb₂Sr₂PrCu₃O₈ compound in which a quasi-2D magnetic order is observed below 7

208 K as evidenced by neutron diffraction study⁵⁰. In the
 209 aforementioned case the ac susceptibility show a saturation
 210 below the ordering temperature rather than a pro-
 211 nounced drop while the differential exhibit a minimum at
 212 the ordering temperature. In our case there is no clear
 213 minimum of the differential curve, yet it would be nec-
 214 essary to perform a neutron diffraction measurement in
 215 order to confirm or deny the presence of long-range mag-
 216 netic order below the T^* .

217 In contrast with CeNiC_2 and NdNiC_2 , PrNiC_2 does
 218 not reveal any clear magnetic transition. Since the three
 219 compounds are chemically similar, the discrepancy arises
 220 likely from the difference in the detailed structure of $4f$
 221 energy levels. The ground state of a free Pr^{3+} ion is
 222 ninefold degenerate with total angular momentum $J =$
 223 4. The crystalline electric field (CEF) acting on the Pr^{3+}
 224 removes the degeneracy (either fully or partially), with
 225 the nature of the effect dependent on the point symmetry
 226 of the ion crystallographic position. In the orthorhombic
 227 PrNiC_2 the $2a$ site occupied by a Pr atom has the point
 228 symmetry group $mm2$. For such relatively low symmetry
 229 one would expect a complete uplifting of the ground state
 230 degeneracy, yielding a nonmagnetic configuration with 9
 231 separated singlet states similarly as in PrNi_2Al_5 ⁵¹. Note
 232 however that in the case of exchange interaction energy
 233 exceeding the first CEF excitation, the magnetic order
 234 may appear due to the intermixing of higher energy states
 235 into a ground state with higher degeneracy⁵². Such situ-
 236 ation occurs in the orthorhombic PrNiGe_2 compound
 237 crystallizing in the CeNiSi_2 -type structure (related to
 238 CeNiC_2) in which the Pr^{3+} ion position has the same
 239 point symmetry as in PrNiC_2 , yet the material reveals
 240 ferromagnetic (FM) ordering at $T_C = 13$ K^{52,53}.

241 Figure 5a, b and c, shows the thermal dependencies
 242 of electrical resistivity (ρ_{xx}) measured without and with
 243 applied magnetic field (9 T), for CeNiC_2 , PrNiC_2 and
 244 NdNiC_2 respectively. At high temperatures, all the com-
 245 pounds exhibit typical metallic behavior with resistivity
 246 decreasing with temperature lowering. Upon cooling, ρ_{xx}
 247 of both PrNiC_2 and NdNiC_2 show the anomalies pro-
 248 nounced by a minimum followed by a hump. This metal-
 249 metal transition is a typical signature of the charge den-
 250 sity wave state with incomplete Fermi surface nesting,
 251 characteristic for quasi-2D materials⁵⁴. The temperature
 252 of this anomaly corresponds to the Peierls temperature
 253 ($T_P = 121$ K for NdNiC_2 and $T_P = 89$ K for PrNiC_2)
 254 established by X-ray diffuse scattering²⁸. In contrast
 255 to that, no CDW-like anomaly is observed in the third
 256 compound, CeNiC_2 . At the magnetic crossover temper-
 257 atures, all three curves exhibit a decrease in resistivity,
 258 shown closer in the insets of Figure 5. This downturn is
 259 visibly sharper for the antiferromagnetic ground states of
 260 NdNiC_2 and CeNiC_2 than in the case of PrNiC_2 , where
 261 instead of a long range of magnetic ordering, one observes
 262 a small magnetic anomaly at T^* .

263 Although the anomalies in the zero field resistivity
 264 have been reported beforehand²⁷, the influence of mag-
 265 netic field on transport properties, up to now, has been

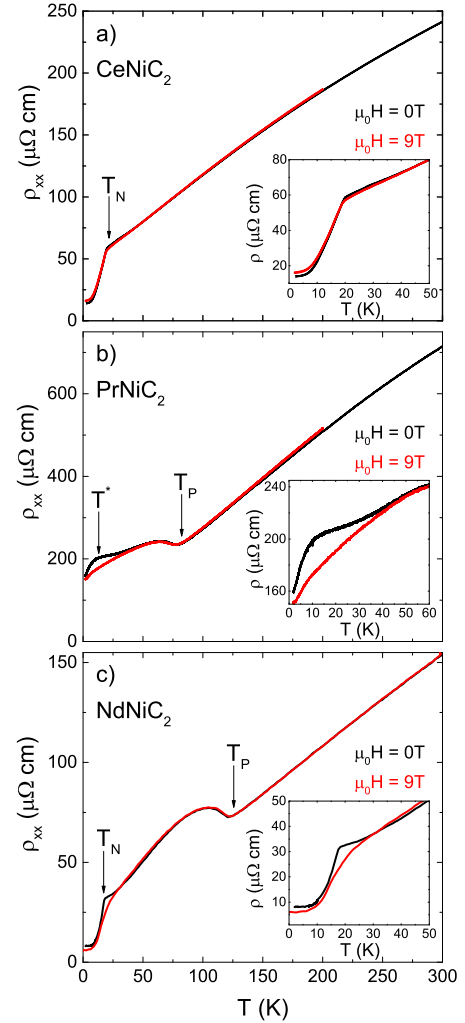


FIG. 5. Resistivity of a) CeNiC_2 , b) PrNiC_2 , c) NdNiC_2 , measured without (black color) and with (red color) applied magnetic field of 9 T. Arrows indicate characteristic temperatures: T_P - Peierls temperature for NdNiC_2 and PrNiC_2 , T_N Néel temperature for CeNiC_2 and NdNiC_2 , and T^* - magnetic anomaly temperature in PrNiC_2 . Insets: Expanded view of the vicinity of the magnetic ordering (anomaly) temperature.

266 studied solely for the Nd-bearing compound^{28,55}. Elec-
 267 trical resistivity measured in the presence of a magnetic
 268 field of $\mu_0H = 9$ T is shown as a red line in Figure 5, a b
 269 and c. The influence of magnetic field on ρ_{xx} in the high
 270 temperature metallic state of each compound is negligi-
 271 bly small. In CeNiC_2 , this behavior is present down to
 272 the vicinity of T_N , where the magnetic field weakly mod-
 273 ifies the resistivity. This is in contrast to the features
 274 seen in the two compounds exhibiting the charge density
 275 waves; in NdNiC_2 one observes a notable decrease in re-
 276 sistance with magnetic field at $T \rightarrow T_N$. In PrNiC_2 the
 277 onset of the negative magnetoresistance can be observed

at $T \approx 60$ K, much closer to T_P than in NdNiC_2 . To investigate further the impact of $\mu_0 H$ on transport properties of studied compounds we have performed the field sweeps at constant temperatures.

The magnetic field dependence of magnetoresistance ($\text{MR} = \frac{\rho(H) - \rho_0}{\rho_0}$, where ρ_0 is the zero field resistivity) of CeNiC_2 is depicted in Figure 6a. At $T > T_N$, MR is weak and negative (resistivity decreases by a maximum of 3%). Below this temperature, the magnetoresistance changes its sign and magnitude. This is a typical picture of the modification of the scattering rate in the vicinity of the magnetic ordering temperature^{56–58}; above T_N the reduction of resistance can be attributed to the field induced ordering of the local magnetic moments, resulting in the quenching of the spin fluctuations and effectively a decrease of the related scattering mechanism. On the other side of the transition, below T_N , the magnetic field induces a partial reorientation of the local spins and perturbs the antiferromagnetic order, which results in the increase of the scattering rate and, consequently, of the electrical resistance.

Figure 6b shows the magnetic field dependence of magnetoresistance of PrNiC_2 . One can notice that, in the charge density wave state, MR is dominated by the negative component which rises as temperature decreases down to T^* . Below this temperature limit, the negative MR decreases and finally at $T = 2$ K a positive term can be observed at low magnetic field. This positive MR component can originate from an onset of another magnetic-like transition at lower temperatures or from the light carriers related to the small Fermi surface pockets that can be opened in the FS due to imperfect nesting. A complementary experiment, such as ARPES spectroscopy, neutron diffraction or magnetotransport measurements performed at temperatures below 1.9 K and higher field would be required to clarify this point. Figure 6c shows the magnetic field dependence of resistivity of NdNiC_2 . Due to the rich variety of positive and negative MR components seen in this compound, we find it more clear to use the $\rho_{xx}(H)$ instead of $\text{MR}(H)$ for discussion of the magnetotransport properties in NdNiC_2 . At 30 K, one observes an onset of the negative magnetoresistance term, which becomes stronger as temperature decreases. Below T_N , the resistivity firstly rises with magnetic field and after reaching the maximum, the ρ_{xx} decreases again. The position of the resistivity maximum at various temperatures below T_N corresponds to the magnetic field induced ferromagnetic transition according to the H - T phase diagram of NdNiC_2 constructed for a single crystal⁴³. Below 14 K, one observes an additional kink (marked in Fig. 6 by arrows) on the decreasing side of resistance. This can be attributed to the intermediate magnetic phase separating the AFM and FM orders at this temperature range. In addition, one can notice that at the lowest temperatures the resistivity saturates at high magnetic fields. The negative magnetoresistance in NdNiC_2 has been attributed^{28,55} both to the suppression of spin disorder scattering and to the destruction

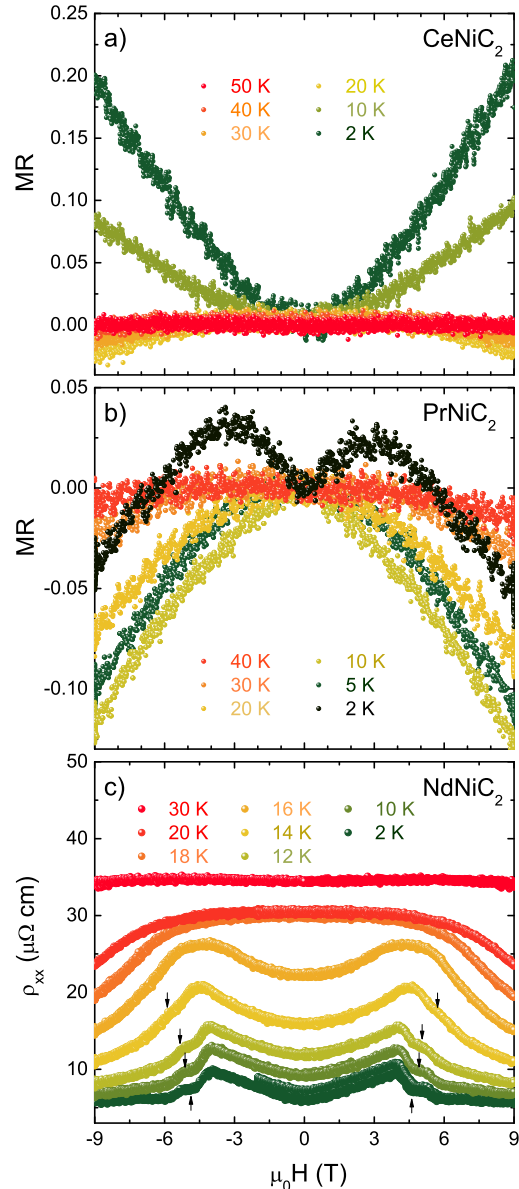


FIG. 6. Magnetotransport properties of $R\text{NiC}_2$. All the measurements have been performed at constant temperature. a) Magnetoresistance in CeNiC_2 as a function of magnetic field, b) Magnetic field dependence of magnetoresistance in PrNiC_2 , c) Resistivity of NdNiC_2 as a function of magnetic field. For better clarity, for this compound we show the ρ_{xx} instead of MR. Arrows indicate the kinks attributed to a metamagnetic phase separating the FM and AFM orders.

of the charge density wave as seen in the isostructural, albeit ferromagnetic compound, SmNiC_2 in which the relevance of the CDW suppression has been confirmed by the X-ray diffuse scattering experiment performed in magnetic field^{59,60}.

An interesting observation is the irreversible behavior

of the electrical resistivity at low temperatures. In order to prove that this effect is not an artifact caused by unstable electrical contacts and is intrinsic to the sample, we have repeated the measurement at lower temperatures. Firstly the sample was warmed up to 40 K, far above the magnetic ordering temperature ($T_N = 17$ K). Next, we have cooled the sample with zero applied field, and stabilized the temperature before activating the magnet. The magnetic field was swept initially to 2 T, to avoid crossing the AFM-FM transition. Then, the magnetic field was swept and reached -9 T (9 T applied in the adverse direction). Afterwards, we performed the final sweep and continuously reversed the direction of the magnetic field to 9 T. The whole procedure was repeated for each scan in order to remove any magnetic memory from the sample. In Figure 7 we show the results of the field sweeps at the selected temperatures. The resistivity measured at $T = 14$ K (Figure 7a) is reversible with $\mu_0 H$. At $T = 10$ K (Figure 7b) one can notice a small irreversibility of ρ_{xx} , which becomes more pronounced at $T = 8$ K, as depicted in Figure 7c. When the magnetic field is increased to 2 T and then swept to 0, the resistivity returns to the zero-field cooled value of ρ_0 . In these conditions, the sample remains in the AFM state. However, the application of a magnetic field exceeding the limit of 4 T, at which the FM order is induced in the sample, prevents the resistance from returning to the original ρ_0 . Further magnetic field sweeps do not induce any irreversible transitions and the resistivity returns to the new value of ρ_0^* when the field is reduced back to 0. Figure 7d compares the result of a field sweep of the sample cooled to 2 K in ZFC condition and the ρ_{xx} of the same sample, which previously experienced the transformation to the FM state at $T = 5$ K (inset). The irreversible behavior is clearly visible in the former case, while in the latter one the resistivity returns to the initial value. This shows that the resistance of NdNiC₂ depends not only on temperature, applied magnetic field or the type of magnetic ordering present in the sample at these conditions, but also on the magnetic history of the sample and this metastable effect is clearly associated with the AFM-FM transition. Previous reports on the magnetoresistance of NdNiC₂^{28,55} have not mentioned the irreversible phase transition, probably because this weak crossover could be easily overlooked, since once the sample experiences the high magnetic field at temperature below 12 K it remains in the metastable state and the irreversibility is no longer observable until the sample is reheated and cooled down again. One plausible scenario to explain this irreversible effect is the magnetoplastic lattice deformation induced by the ferromagnetic transition. Note that even a small lattice transformation and a consequent Fermi surface modification can substantially impact the nesting conditions and this can lead to the quasi-permanent suppression of CDW.

The BCS approach predicts the negative magnetoresistance in CDW systems to originate from the Zeeman splitting of the conduction bands⁶¹ which results in re-

duction of the pairing interactions and degradation of nesting properties. This term has been found to originate both from orbital effects and from local spins producing stronger magnetic moments. For magnetic fields $\mu_B H \ll \Delta_{CDW}$, the Zeeman magnetoresistance term is expressed⁸ by Equation 3:

$$MR = \frac{\rho(H) - \rho_0}{\rho_0} = -\frac{1}{2} \left(\frac{\mu_B H}{k_B T} \right)^2 + 0 \left(\frac{\mu_B H}{k_B T} \right)^4 \quad (3)$$

The Figure 8a shows the magnetoresistance of NdNiC₂ above T_N as a function of $\frac{1}{2} \left(\frac{\mu_B H}{k_B T} \right)^2$. The plots do not converge into a single straight line. This is not surprising, since this temperature interval corresponds to the onset of the field induced magnetic ordering. This can lead either to the previously suggested CDW suppression, stronger than predicted by Equation 3 or to the reduction of the spin scattering, which also results in negative magnetoresistance as in CeNiC₂. The comparison of the strength of the negative magnetoresistance in NdNiC₂ and CeNiC₂ in the vicinity of T_N can also be a useful guide. In the former compound, showing the Peierls instability, MR reaches -40 % which is an order of magnitude larger than in the latter one, in which the CDW is absent. This suggests that, the negative magnetoresistance in NdNiC₂ originates, at least partially, from the suppression of the CDW state.

The negative MR in PrNiC₂ reaches a maximum of 12%, which although is visibly weaker than in NdNiC₂, still exceeds the value found in CeNiC₂. This, similar to the case of NdNiC₂, suggests that the decrease of resistance in magnetic field originates from the suppression of the CDW. To verify this hypothesis, we have scaled the magnetoresistance in PrNiC₂ with Equation 3, as shown in Figure 8 b. At $T > 20$ K the PrNiC₂ can be qualitatively described by the Zeeman term; the MR plots fall into a single straight line. At lower temperatures, in the vicinity of T_M the negative magnetoresistance is weakened and diverges from this scaling law (as shown in the inset of Figure 8b). The curve obtained for $T = 10$ K is a boundary of the relevance of the Equation 3. At $\frac{1}{2} \left(\frac{\mu_B H}{k_B T} \right)^2 \approx 0.02$, which corresponds to $\mu_B H = 6$ T at this temperature, the magnetoresistance plot diverges from the Zeeman scaling and starts decreasing. We find that, to apply Equation 3 one has to use the prefactor of approximately 1.4. In other CDW materials this coefficient is usually smaller than unity. The key examples are Li_{0.9}Mo₆O₁₇⁶² or organic compounds such as (Per)₂Pt(mnt)₂⁶³⁻⁶⁶ in which the existence of weakly magnetic chains ramps this magnetoresistance prefactor in comparison with (Per)₂Au(mnt)₂^{67,68} showing a non-magnetic character. On the other hand, the value we found is significantly lower than the factor of ≈ 30 found in GdNiC₂⁴⁹, where the presence of strong local magnetic moments amplifies the internal magnetic field much more effectively than in PrNiC₂, showing no clear long range magnetic ordering.



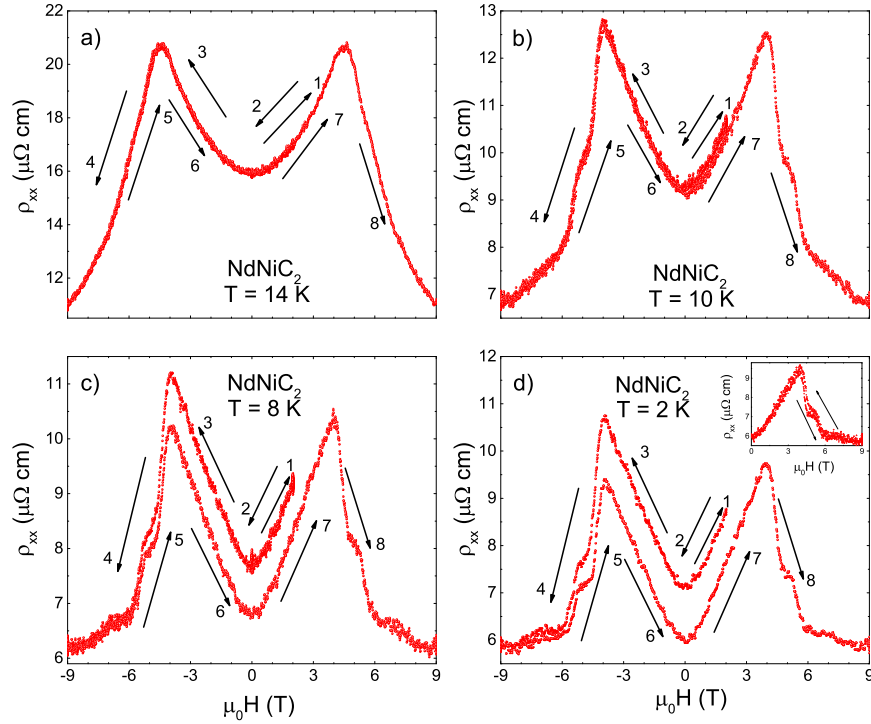


FIG. 7. Resistivity of NdNiC₂ measured at selected temperatures. After each field sweep data collection at constant temperature, the sample was warmed up to 40 K in zero magnetic field to remove the magnetic memory of the material. Arrows and numbers show the direction of field sweeps. a) $T = 14$ K, b) $T = 10$ K, c) $T = 8$ K, d) $T = 2$ K. Inset: Resistivity at $T = 2$ K of the same sample of NdNiC₂, however previously subjected to the magnetic field of 9 T at $T = 5$ K.

454 Due to polycrystalline nature of our samples, we are
 455 unable to perform the X-ray diffuse scattering experi-
 456 ment to follow the intensity and position of the satellite
 457 reflections at various temperature and magnetic field. In-
 458 stead, to investigate the suppression of the charge den-
 459 sity waves state by magnetic field, we have conducted
 460 the Hall effect measurements, which can be used as a di-
 461 rect probe for electronic carrier concentration. Figure 9a
 462 shows the thermal dependence of Hall resistivity (ρ_{xy})
 463 in NdNiC₂. The sign of the measured Hall resistance
 464 is negative, opposite to the results reported recently⁵⁵.
 465 To clarify this point, we have repeated the measurement
 466 with a reference sample of Cu foil, which shows a nega-
 467 tive Hall signal in the same contact geometry. This con-
 468 firms the relevance of the negative sign of ρ_{xy} in NdNiC₂.
 469 At $T > T_P$, the Hall signal is almost independent of
 470 temperature. At the Peierls temperature one observes
 471 a downturn of $\rho_{xy}(T)$ (and increase of $|\rho_{xy}|$), which is
 472 a typical signature of the opening of the CDW bandgap
 473 and condensation of electronic carriers^{69,70}. Upon fur-
 474 ther cooling, the Hall resistivity decreases until it reaches
 475 a minimum followed by a prominent increase of ρ_{xy} (and
 476 decrease of $|\rho_{xy}|$), which grows even higher than for tem-
 477 peratures above T_P .

478 This increase of ρ_{xy} in proximity of the magnetic or-
 479 dering temperature observed in SmNiC₂⁷¹ and NdNiC₂⁵⁵

480 has been attributed to the destruction of CDW and a
 481 concomitant release of previously condensed carriers. Al-
 482 though the CDW suppression by magnetic field appears
 483 to be quite a possible scenario, this mechanism itself is
 484 not sufficient to explain the features observed as $T \rightarrow T_N$,
 485 especially considering that the low temperature $|\rho_{xy}|$ is
 486 lower than the value found for $T > T_P$. This could lead
 487 to a misleading suggestion that the carrier concentra-
 488 tion below T_N exceeds the high temperature normal state
 489 value. To avoid the oversimplification, in a material ex-
 490 hibiting magnetic ordering, one has to consider two com-
 491 ponents of the Hall resistance⁷²:

$$\rho_{xy} = R_0\mu_0H + 4\pi R_S M \quad (4)$$

492 The R_0 in Equation 4 is the ordinary Hall coefficient
 493 which, in a single band model, is inversely proportional
 494 to the carrier concentration. R_S denotes the anomalous
 495 Hall coefficient associated with side jump and skew scat-
 496 tering. To obtain the more clear evidence of the par-
 497 tial CDW destruction in NdNiC₂, we complement the
 498 previous Hall effect study⁵⁵ of this compound in re-
 499 gard to the anomalous component of the Hall signal.
 500 We also present the results of the same experiment for
 501 CeNiC₂ and PrNiC₂ which similarly to magnetoresis-
 502 tance in these two compounds have not been reported

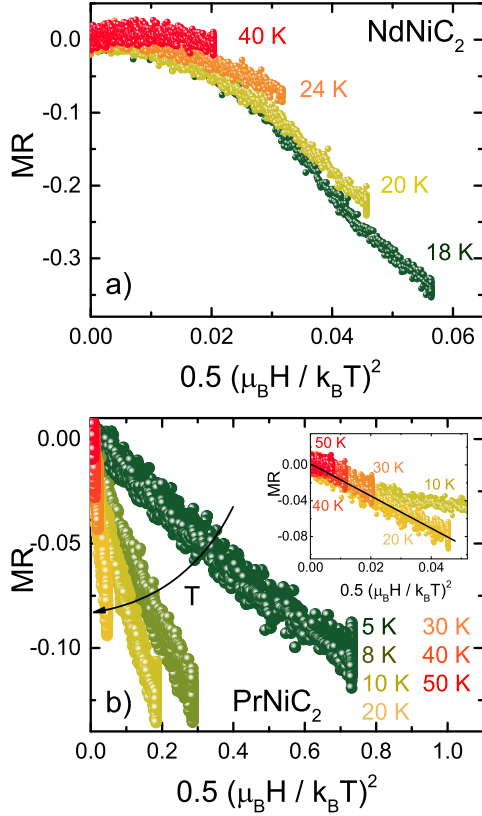


FIG. 8. Scaling of magnetoresistance in PrNiC_2 with Equation 3. Inset: Expanded view of the MR scaling for $T \geq 10$ K

503 previously. The separation of normal and anomalous ρ_{xy}
 504 components is not straightforward unless the magnetic
 505 moment saturates with magnetic field which then reduces
 506 the latter one to a constant^{73–76}. Here, no signs of saturation
 507 of $M(T)$ up to an applied field of 14 T for any
 508 of the studied compounds have been found⁷⁷, which pre-
 509 cludes the possibility of the direct extraction of electronic
 510 concentration from ρ_{xy} . Nevertheless we can propose an
 511 alternative road to follow the number of carriers condensed
 512 into the charge density wave state. The idea is
 513 to compare the field dependencies of ρ_{xy} and M with a
 514 special regard for the temperature region, in which mag-
 515 netization follows the linear field dependency. In this
 516 condition the anomalous component contribution is also
 517 linear with field and, for a single band metal, any depart-
 518 ure from the linearity of ρ_{xy} indicates the change of
 519 R_0 which is a measure of electronic concentration.

520 Figure 9b shows the magnetic field dependence of the
 521 Hall resistivity of NdNiC_2 measured at various temper-
 522 atures. At $T \geq 60$ K one cannot find any departure from
 523 linearity for the $\rho_{xy}(H)$. A small nonlinearity can be seen
 524 at 40 K. Upon further cooling, the deviation from linear
 525 variation for $\rho_{xy}(T)$ becomes more pronounced. Com-

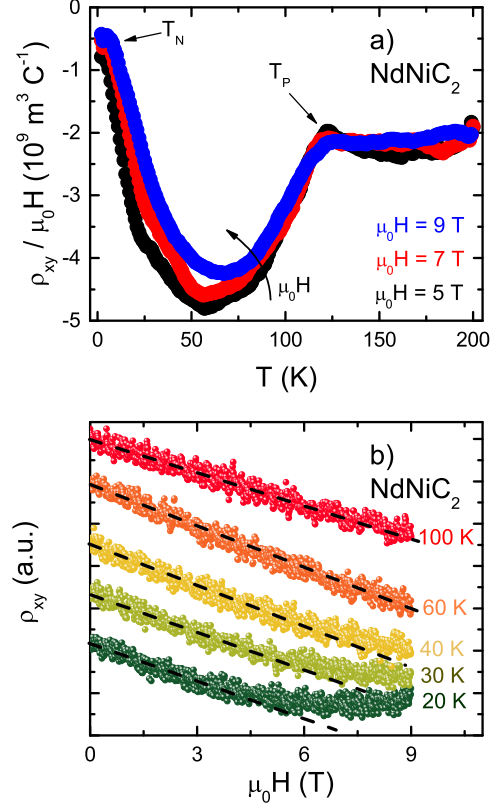


FIG. 9. a) Hall resistivity of NdNiC_2 , divided by magnetic field, measured at various magnetic fields. Arrows indicate the Peierls and Néel temperatures T_P and T_N respectively. b) Hall resistivity of NdNiC_2 as a function of magnetic field. The plots have been shifted horizontally to improve data reading.

526 paring this result with magnetization data for NdNiC_2
 527 (Fig. 2d), which shows linear $M(H)$ dependence at $T \geq$
 528 20 K one can deduce that, in this temperature range,
 529 the non-linearity of $\rho_{xy}(H)$ can be safely attributed to
 530 the increase in electronic concentration. This indicates
 531 that, the release of previously CDW condensed carriers
 532 is, next to the anomalous Hall component, responsible
 533 for the increase of ρ_{xy} as temperature is lowered to the
 534 vicinity of T_N . Here we emphasize that, since we were
 535 unable to observe the saturation of $M(H)$ we are unable
 536 to separate the normal and anomalous components of the
 537 Hall resistivity for $T \leq 20$ K, where both ρ_{xy} and M are
 538 non-linear functions of $\mu_0 H$. The thermal dependence of
 539 Hall resistance of PrNiC_2 depicted in Figure 10a exhibits
 540 some similarities to the case of NdNiC_2 . A significant
 541 downturn of ρ_{xy} below T_P concomitant with an increase
 542 of resistivity (Figure 5c) due to the condensation of the
 543 electronic carriers is observed at T_P . Upon further cool-
 544 ing, the Hall resistivity continues to decrease and does
 545 not simply saturate at $\frac{T_P}{2}$, where the electronic gap is

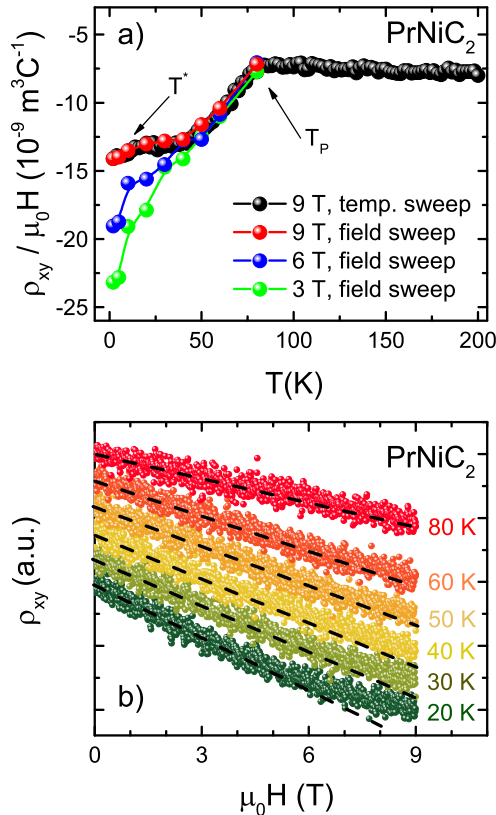


FIG. 10. a) Hall resistivity of PrNiC₂, divided by magnetic field, black points show the data collected from the temperature sweep at constant magnetic field of 9 T. Red, blue and green points show the data collected from the field sweeps at constant temperature. Arrows indicate the Peierls and magnetic transition temperatures T_P and T^* respectively. Solid lines are the guide for the eye. b) Hall resistivity of PrNiC₂ as a function of magnetic field. The plots have been shifted horizontally to improve data reading. Dashed lines show the low field linear dependencies of $\rho_{xy}(H)$ expanded to the high field regime.

547 expected to be fully open. This behavior is consistent
 548 with the non-BCS thermal dependence of the satellite
 549 reflections intensity²⁸ suggesting that the nesting vector
 550 adjusts to the FS evolution. In contrast to NdNiC₂, no
 551 significant upturn of ρ_{xy} is observed as T approaches the
 552 magnetic ordering temperature. Contrarily, below T^* the
 553 Hall resistivity starts to decrease again. This observa-
 554 tion is in agreement with the behavior of the intensity
 555 of the CDW satellite reflections²⁸, which show a sud-
 556 den increase upon crossing T^* . Below $T \approx 60$ K, corre-
 557 sponding to the onset of negative magnetoresistance, the
 558 $\rho_{xy}(T)$ curves obtained at different magnetic fields do
 559 not converge. The application of stronger magnetic field
 560 drives the thermal dependence of ρ_{xy} towards more pos-
 561 itive values, in comparison to the data obtained at lower

562 H . Similar to NdNiC₂, this can be attributed to the pos-
 563 itive anomalous Hall component growing as the magneti-
 564 zation increases or to the partial suppression of the CDW
 565 and increase of the electronic concentration. It shall be
 566 noted that, the strength of the ρ_{xy} downturn below T^*
 567 is sufficient to overcome the anomalous term driving the
 568 Hall resistivity towards more positive values. Note that,
 569 the strength of the anomalous Hall signal in PrNiC₂ is
 570 expected to parallel the scale of NdNiC₂, since the val-
 571 ues of magnetization of both compounds are comparable.
 572 To explore this effect further, we have conducted $\rho_{xy}(H)$
 573 measurements for PrNiC₂. As shown in Figure 10b, the
 574 non-linearity of the Hall resistivity plotted versus $\mu_0 H$
 575 can be observed in this compound as well. The devia-
 576 tion from linearity, initially barely observable for $T = 50$
 577 K becomes stronger at lower temperatures. Here, how-
 578 ever, we cannot follow the same analysis as for the case of
 579 NdNiC₂, due to the fact that for temperatures lower than
 580 60 K the magnetization does not follow a linear relation-
 581 ship with $\mu_0 H$. Therefore, the two normal and anoma-
 582 lous ingredients of the Hall resistivity in PrNiC₂ cannot
 583 be unambiguously separated. Nevertheless, the down-
 584 turn of ρ_{xy} at T^* strongly suggests the enhancement of
 585 the CDW state, although the magnetoresistance above
 586 T^* shows some signatures of the partial suppression of
 587 the Peierls instability. This can be explained in terms
 588 of the lattice transformation accompanying the magnetic
 589 anomaly modifying the Fermi surface, which triggers the
 590 nesting of another FS part when the CDW vector ad-
 591 justs to band structure evolution. One cannot however
 592 exclude an alternative scenario, in which the enhance-
 593 ment of the Fermi surface nesting can be seen as a driving
 594 force for the magnetic anomaly. Since the magnetic prop-
 595 erties are related to the free electron density via RKKY
 596 interactions, it is not unreasonable to expect the conden-
 597 sation of the electronic carriers at T^* to modify of the
 598 magnetic character of PrNiC₂. The high resolution X-
 599 ray and neutron diffraction experiment performed with
 600 a single crystal of PrNiC₂ will be required to clarify this
 601 point.

602 The thermal dependence of Hall resistivity in CeNiC₂,
 603 shown in Figure 11a shows no signatures of electronic
 604 condensation. This is in agreement with transport prop-
 605 erties in which no anomalies similar to those found in
 606 NdNiC₂ and PrNiC₂ are observed and confirms the ab-
 607 sence of the Peierls instability in CeNiC₂. From the clear
 608 correlation between the thermal dependence of ρ_{xy} and
 609 magnetization (see Figure 11b), one can conclude, that
 610 the anomalous component is the dominant ingredient of
 611 the Hall effect in this compound, while the normal Hall
 612 coefficient is expected to remain temperature independ-
 613 ent. The observation of the increase of ρ_{xy} as $T \rightarrow T_N$
 614 in CeNiC₂, where the absence of the CDW has been em-
 615 phasized, implies that the anomalous Hall component is
 616 essential to describe the ρ_{xy} in NdNiC₂ and PrNiC₂.

617 To explore the observed transitions further, we have
 618 studied the thermal and magnetic field dependencies of
 619 specific heat (C_p). Previously the $C_p(T, H)$ has been

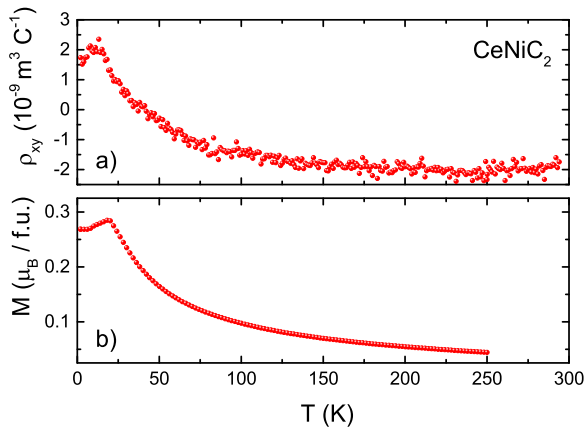


FIG. 11. Hall resistivity in CeNiC_2 as a function of temperature (a) compared with magnetization (b) of the same compound

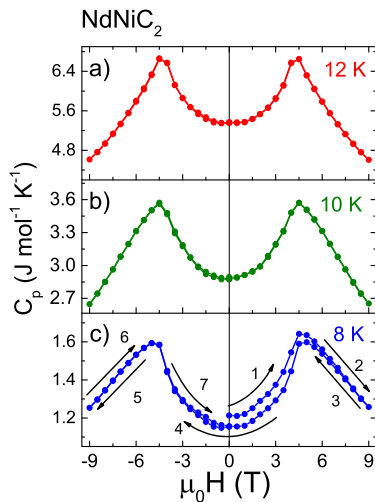


FIG. 12. Specific heat of NdNiC_2 as a function of magnetic field measured at a) $T = 12$ K, b) $T = 10$ K, and c) $T = 8$ K. Arrows and numbers show the direction of the magnetic field sweeps. At each temperature step the sample was first heated to 40 K, well above the magnetic transition temperature $T_N = 17$ K, held for a few minutes and then cooled to the target temperature with no applied magnetic field. After stabilizing the temperature, the magnetic field was first increased to 9 T, then decreased to -9 T and swept to 0 T. At 8 K an irreversible behavior is clearly seen - during the first field sweep the specific heat below 4.5 T is higher than for the second sweep from +9 to -9 T, indicating the formation of a field-induced metastable phase, which is also observed in transport measurements.

620 successfully used to construct the phase diagram for
 621 GdNiC_2 ⁴⁹. Figure 13 shows a specific heat map (a) and
 622 the heat capacity of the polycrystalline CeNiC_2 (b) plot-
 623 ted as a function of temperature, under various magnetic
 624 fields. In the results we can observe a few anomalies.
 625 The largest one is seen at about 19 K and is almost un-

626 affected by the applied magnetic fields up to 9 T. The
 627 second anomaly is less pronounced and the temperature
 628 of its occurrence varies with the applied magnetic field
 629 from 11 K in 0 T to 9.5 K in 9 T. The existence of the fea-
 630 tures anomalies are in agreement with magnetization and
 631 transport results. Another anomaly, previously reported
 632 by Motoya et al.⁴⁶, seen at 2 K is magnetic field depen-
 633 dent. A minor jump around 30 K is likely connected with
 634 the CeC_2 impurity phase⁴⁴, as suggested from magnetic
 635 susceptibility data.

636 The broad hump seen in PrNiC_2 (Fig. 13 c and d) is a
 637 Schottky anomaly originating from multiple energy lev-
 638 els of the Pr^{3+} ion subject to the CEF splitting. Due to
 639 the complicated energy level structure the specific heat
 640 data could not be reliably fitted in order to extract the
 641 level splitting energies. The anomaly is slightly shifted
 642 towards higher temperature by applied magnetic field as
 643 seen in Figure 13 c and d, which is caused by the Zeeman
 644 effect, as seen in many f -electron systems (see eg.⁷⁸⁻⁸⁰).
 645 No clear anomaly is seen around T^* corresponding both
 646 to the drop in the Hall resistivity and the upturn of sus-
 647 ceptibility. This may suggest that the alleged transition
 648 involves predominantly the change of electronic structure
 649 with little effect on crystal and spin order, which should
 650 result in the appearance of an anomaly in specific heat.
 651 Note that in the $\text{Pb}_2\text{Sr}_2\text{PrCu}_3\text{O}_8$ compound mentioned
 652 before the specific heat anomaly at the transition tem-
 653 perature is weak⁸¹. If such weak anomaly would arise in
 654 PrNiC_2 at the T^* it could be hard to observe on top of
 655 the large Schottky hump.

656 The results of the specific heat measurements for
 657 NdNiC_2 are shown in Fig. 13 e and f. For this com-
 658 pound the specific heat shows a lambda-like anomaly at
 659 T_N , which is weakly affected by the applied magnetic
 660 field up to about 3.0-3.5 T above which a metamagnetic
 661 transition occurs. Above 7 T we can observe the third
 662 anomaly which is probably related to the occurrence of
 663 the transitional phase between AFM and FM.

664 The magnetic field dependence of the specific heat of
 665 NdNiC_2 measured at 12 K, 10 K and 8 K is presented
 666 in Fig. 12. At 8 K the C_p vs. H shows an irreversible
 667 behavior as seen in Figure 12c. The observation of the
 668 irreversibility in both specific heat and electrical resis-
 669 tivity measurements confirms the presence of a magnetic
 670 field-induced metastable state, not reported in previous
 671 studies. Interestingly, the same transition does not re-
 672 sult in the appearance of hysteresis in magnetization, as
 673 seen in the inset of Figure 2. This could be explained
 674 by the insufficient resolution of magnetization measure-
 675 ments performed with the ACMS option. However it is
 676 also possible that the field-induced transition involves a
 677 change of electronic and crystal structures without a sig-
 678 nificant change in magnetic order.

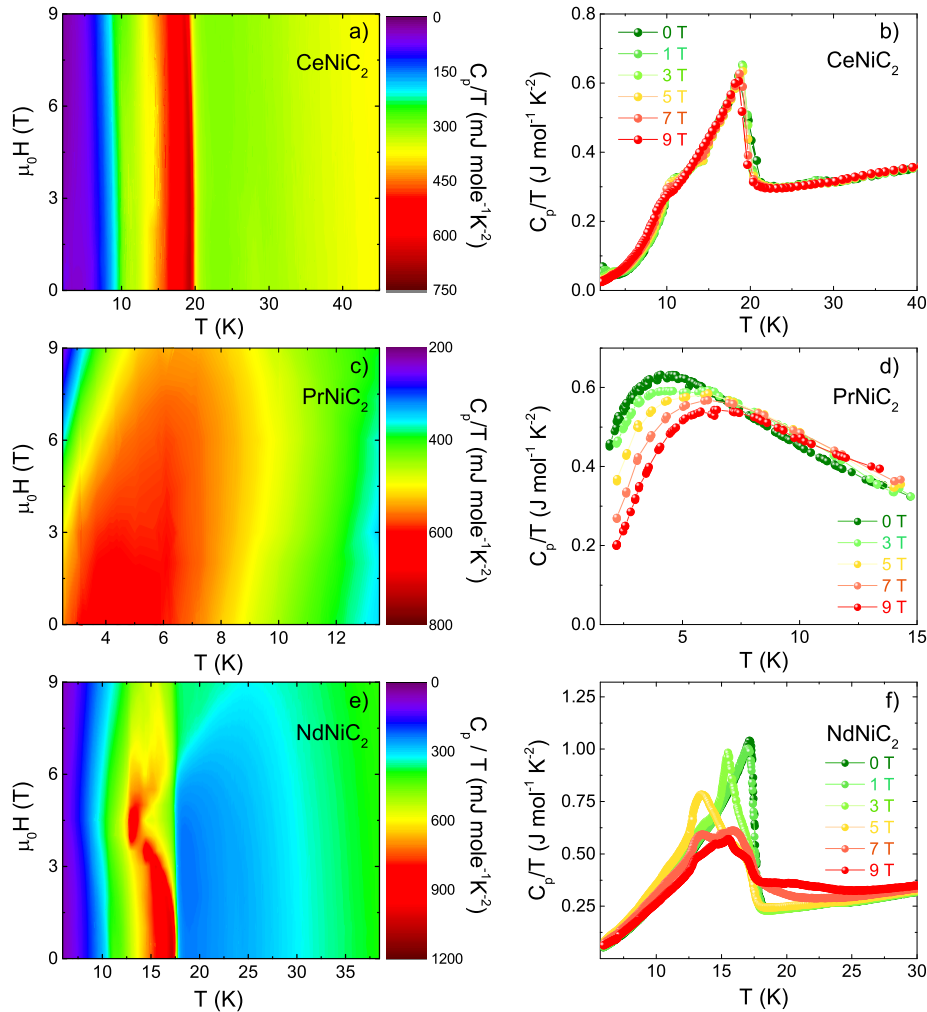


FIG. 13. Panels a) and b) present the specific heat of CeNiC₂ as a function of temperature and magnetic field. The anomaly seen at $T_N = 19$ K does not significantly shift with applied magnetic fields up to 9 T, while the anomalies around 10 and 2 K are suppressed by increasing $\mu_0 H$. Panels c) and d) show the specific heat of PrNiC₂, revealing that the broad hump, attributed to the Schottky anomaly resulting from splitting of the f orbital energy levels is gradually shifted towards higher temperatures by application of a magnetic field due to the Zeeman effect. Panels e) and f) present the specific heat of NdNiC₂. The anomaly at 17 K remains almost unaffected by magnetic fields up to approx. 3 T above which a field-induced magnetic transition takes place, as evidenced by magnetization and transport measurements. At higher fields the specific heat curves develop a complicated structure indicating that the magnetic phase diagram is complex, as previously reported for GdNiC₂⁴⁹.

IV. CONCLUSIONS

In order to explore the interaction between charge density waves and magnetism in the RNiC₂ family, we have compared the physical properties of three isostructural compounds: NdNiC₂, showing both the Peierls instability, PrNiC₂ with the CDW and a magnetic anomaly, and CeNiC₂, showing antiferromagnetic ordering, and the absence of the CDW transition. The weak magnetoresistance in CeNiC₂ is found to originate by the spin fluctuations

accompanying the magnetic transition. Neither transport or Hall effect measurements reveal any signatures of the Peierls instability. Study of the magnetoresistance and the galvanomagnetic properties of NdNiC₂ confirms the partial suppression of charge density waves by magnetic ordering and a further destruction of the Peierls instability at the crossover from the antiferromagnetic to ferromagnetic order. We have also found that this magnetic transformation drives a metastable lattice transformation that can be observed via the mag-



netoresistance and the specific heat measurements. The interplay between magnetism and charge density waves in PrNiC₂ shows more complex character. Although the magnetoresistance data suggest that, the application of magnetic field partially suppresses CDW by Zeeman splitting of the electronic bands, the expansion of the nested region of the Fermi surface at $T^* \approx 8$ K can be observed by a significant downturn of the Hall resistivity, strong enough to overcome the positive Hall signal originating from the anomalous component. This effect seems to be related to the magnetic anomaly⁴³ observed at the same temperature, however the underlying mechanism remains unclear. Tentatively, the interaction between the CDW and magnetic properties of this compound can be described either by the lattice transformation due to the magnetic anomaly, and by the modification of the magnetic ordering via the RKKY interactions influenced by change of the electronic concentration. Further analysis of this effect can be realized by high resolution diffraction experiments on a single crystal.

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