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Non-Fermi-liquid types of behavior associated with a magnetic quantum-critical point in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ single crystals

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The compound SrCo_2As_2 with the body-centered tetragonal ThCr_2Si_2 structure is known to remain paramagnetic down to a temperature $T = 0.05$ K, but inelastic neutron scattering studies have shown that both ferromagnetic (FM) and antiferromagnetic (AFM) fluctuations occur in single crystals. Thus it is of interest to study how the magnetism evolves on doping SrCo_2As_2 . Previous work on polycrystalline samples of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ indicated the development of AFM order for $0 < x \lesssim 0.3$. Here we studied single crystals of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ for $0 < x \leq 1$ and confirmed the occurrence of AFM order which we deduce to have a c -axis helix structure. We also find evidence for an unusual composition-induced magnetic quantum-critical point at $x \approx 0.3$ where non-Fermi-liquid types of behavior were revealed by heat capacity and electrical resistivity measurements at low T . Electron-doped $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ single crystals with compositions $x = 0$ to 0.9 were grown out of self-flux and SrNi_2As_2 single crystals out of Bi flux. The crystals were characterized using single-crystal x-ray diffraction (XRD) at room temperature, and magnetic susceptibility $\chi(H, T)$, isothermal magnetization $M(H, T)$, heat capacity $C_p(H, T)$, and electrical resistivity $\rho(H, T)$ measurements versus applied magnetic field H and T . The XRD studies show that the system undergoes a continuous structural crossover from the uncollapsed-tetragonal (ucT) structure to the collapsed tetragonal (cT) structure with increasing Ni doping. The $\chi(T)$ data show that SrCo_2As_2 exhibits an AFM ground state almost immediately upon Ni doping on the Co site. *Ab-initio* electronic-structure calculations for $x = 0$ and $x = 0.15$ indicate that a flat band with a peak in the density of states just above the Fermi energy is responsible for this initial magnetic-ordering behavior on Ni doping. The AFM ordering is observed in the range $0.013 \leq x < 0.25$ with the ordered moments aligned in the ab plane and with a maximum ordering temperature $T_N = 26.5$ K at $x = 0.10$. The Curie-Weiss-like T dependence of χ in the paramagnetic (PM) state indicates dominant FM interactions. The behavior of the anisotropic susceptibilities below T_N suggests a planar helical magnetic ground state with a composition-dependent pitch based on a local-moment molecular-field-theory model, with FM interactions in the ab plane and weaker AFM interactions along the helix c axis. However, the small ordered (saturation) moments $\sim 0.1 \mu_B$ per transition metal atom, where μ_B is the Bohr magneton, and the values of the Rhodes-Wohlfarth ratio indicate that the magnetism is itinerant. The high-field $M(H)$ isotherms and the low-field $\chi^{-1}(T > T_N)$ data were successfully analyzed within the framework of Takahashi's theory of FM spin fluctuations. The $C_p(T)$ at low T exhibits Fermi-liquid behavior for $0 \leq x \leq 0.15$ whereas an evolution to a logarithmic non-Fermi-liquid (NFL) behavior is found for $x = 0.2$ to 0.3. The logarithmic dependence is suppressed in an applied magnetic field. The low- T $\rho(H = 0, T)$ data show a T^2 dependence for $0 \leq x \leq 0.20$ and a power-law dependence $\rho(H = 0, T) = \rho_0 + AT^n$ with $n < 2$ for $x = 0.20$ and 0.30. The exponent n shows a notable field dependence, suggesting both doping- and magnetic-field-tuned quantum critical phenomena. These low- T NFL types of behavior observed in the C_p and ρ measurements are most evident near the quantum-critical concentration $x \approx 0.3$ at which a $T = 0$ composition-induced transition from the AFM phase to the PM phase occurs.

I. INTRODUCTION

The discovery of high- T_c superconductivity (SC) in iron-based pnictides and chalcogenides motivated many studies of their correlated lattice, electronic, magnetic, and superconducting properties¹⁻⁹. Among such compounds, the class of AT_2X_2 (abbreviated as 122) ternary compounds ($A =$ alkaline earth and lanthanides), which have the body-centered tetragonal ThCr_2Si_2 -type crystal structure with space group $I4/mmm$, have been well studied. By electron or hole doping via chemical substi-

tution which modify the lattice, electronic, and/or magnetic degrees of freedom, the AFe_2As_2 ($A = \text{Sr}, \text{Ca}, \text{Ba}, \text{and Eu}$) compounds were found to exhibit a variety of ground states including different crystallographic structures, magnetically-ordered states or SC. This in turn prompted the search for novel physical properties with different transition-metal-based 122-type compounds such as described in Refs.¹⁰⁻¹⁹.

These tetragonal compounds can often be categorized as having either a collapsed-tetragonal (cT) or uncollapsed-tetragonal (ucT) structure. The structure

is still ThCr_2Si_2 -type, the difference being reflected in the tetragonal c/a ratio. As shown in Fig. 1 below, the structure of a 122-type AM_2Pn_2 compound is comprised of $Pn - M - Pn$ layers separated by the A atoms, where A is usually an alkali-metal or alkaline-earth atom, M is a transition metal such as Fe, Co, Ni, Cu, and Pn is a pnictogen atom such as P or As, with M tetrahedrally coordinated by Pn atoms. The $Pn - M - Pn$ layers are stacked along the tetragonal c axis with the Pn atoms of one $Pn - M - Pn$ layer directly above or below the Pn atoms of an adjacent layer. If there is no covalent bonding between the Pn atoms between layers, the c/a ratio is “large” (ucT structure, more two-dimensional), whereas if such bonding occurs, the c/a ratio is “small” (cT structure, more three-dimensional). In practice, the c/a ratio of different compounds form a continuum between these two limits. A continuum is also found versus composition in various systems including $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ as seen later in Sec. III. For a review, see Sec. VIII of Ref.²⁰.

The Co-based pnictides $A\text{Co}_2\text{As}_2$ ($A = \text{Ca}, \text{Sr}, \text{Ba}$, and Eu) have a variety of magnetic properties. For example EuCo_2As_2 and EuCo_2P_2 exhibit Eu ($4f$) local-moment AFM magnetic ordering with the ucT structure at ambient pressure^{21,22}. These compounds show changes from the ucT to the cT structure under pressure, resulting in a change from Eu ($4f$) local-moment ordering to Co ($3d$) itinerant magnetic ordering^{23–25}. On the other hand, $\text{CaCo}_{2-y}\text{As}_2$ reveals a different magnetic ordering which has a cT structure and undergoes an itinerant A-type AFM ordering where the Co moments are FM aligned along the tetragonal c axis within a Co layer in the ab plane and are AFM aligned between planes along the c axis. Thus the magnetism is dominated by strong ab -plane FM interactions compared to weak AFM interlayer interactions along the c axis²⁶. Inelastic neutron studies revealed that $\text{CaCo}_{2-y}\text{As}_2$ exhibits perfect magnetic frustration, rarely seen in itinerant magnets, between nearest and next-nearest neighbor magnetic interactions. This means that stripe-type AFM and itinerant FM ground states compete which is unique among quasi-two-dimensional AFMs²⁷. In contrast to metallic $\text{CaCo}_{2-y}\text{As}_2$, metallic SrCo_2As_2 and BaCo_2As_2 have the ucT structure with no long-range magnetic ordering and exhibit Stoner-enhanced paramagnetism^{10,28,29}. Furthermore, inelastic neutron scattering measurements on SrCo_2As_2 revealed strong stripe-type AFM correlations at high energies as seen in iron based superconductors, a key factor for inducing SC at high temperatures^{30,31}, whereas NMR and inelastic neutron scattering measurements reveal strong FM correlations in addition to AFM correlations^{32–34}, suggesting a reason that SrCo_2As_2 does not exhibit SC.

The growth of pseudoternary crystals having different properties can also reveal additional interesting physics. For example, crystals of the mixed system $\text{Ca}_{1-x}\text{Sr}_x\text{Co}_{2-y}\text{As}_2$ exhibit a continuous composition-induced crossover in the itinerant magnetic properties between the above-noted A-type AFM I phase ($0 \leq x \leq$

0.2) with c -axis moment alignment to an AFM II phase ($x = 0.40$ and 0.45) with the ordered moments aligned in the ab plane, followed by a paramagnetic (PM) region for $x \geq 0.52$ ^{35,36}.

The ground state of the 122-type pseudoternary series $\text{Sr}_{1-x}\text{Ca}_x\text{Co}_2\text{P}_2$ changes from a nearly-FM Fermi liquid to AFM, then to FM like, and finally back to AFM upon Ca substitution³⁷. These changes in the magnetism are accompanied by a continuous structural crossover from the ucT ($x = 0$) to the cT structure ($x = 1$) that is correlated with the variation in the interlayer P-P bonding distance. It was inferred that the valence state of the transition metal and the P-P interlayer distance are correlated and that the charge redistribution in the Co_2P_2 layers during the ucT to cT transition changes the magnetic ground state. Thus the system $\text{SrCo}_2(\text{Ge}_{1-x}\text{P}_x)_2$ develops weak itinerant FM during the course of P-P dimer breaking and a quantum critical point (QCP) is observed at the onset of the FM phase although both end members SrCo_2P_2 (ucT) and SrCo_2Ge_2 (cT) are PM metals. Here, the FM and QCP are not induced by a simple electron doping effect giving rise to a valence change of the Co atoms, but rather by the breaking of the dimer. When the dimer is fully broken, the FM state is not present and the PM state appears.

On the other hand, $\text{CaCo}_2(\text{Ge}_{1-x}\text{P}_x)_2$ compounds show nonmagnetic to AFM transitions without FM in the intermediate composition region while retaining fully intact X-X dimers ($X = \text{Ge}/\text{P}$) over the whole x doping range³⁸. In contrast, throughout the $\text{BaCo}_2(\text{Ge}_{1-x}\text{P}_x)_2$ system only a nonmagnetic ground state is observed in which the X-X dimer is fully broken (there is no X-X bond) because of the large size of the Ba^{+2} ions³⁸.

The above studies on mixed Co122 systems illustrate the importance of pnictogen Pn - Pn interlayer bonding on the electronic states of Co- Pn layers. Unlike local-moment magnetism, itinerant magnetism originates from the properties of band electrons near the Fermi surface. As a result, the type of magnetic order is determined by the wave vector \mathbf{q} at which the the wave-vector dependent susceptibility $\chi(\mathbf{q})$ has a peak. For FM ordering, $\mathbf{q} = 0$ and the presence or absence of FM long-range ordering is determined by the value of the density of states (DOS) at the Fermi energy E_F . In $A\text{Co}_2\text{As}_2$ ($A = \text{Ba}, \text{Sr}, \text{Ca}$), a sharp peak in the DOS at E_F arises from a Co $d_{x^2-y^2}$ flat band³⁹. A sufficiently large value leads to FM ordering according to Stoner theory. For CaCo_2As_2 , the peak in the DOS is very close to E_F whereas it is about 35 meV above the Fermi level in BaCo_2As_2 and SrCo_2As_2 ³⁹, suggesting that CaCo_2As_2 would have much stronger low-energy FM spin fluctuations than BaCo_2As_2 and SrCo_2As_2 . This is evidently the reason why only CaCo_2As_2 has an A-type AFM transition, where the ordered moments are aligned ferromagnetically within the Co layers and antiferromagnetically between layers, with the AFM interactions much weaker than the FM interactions, whereas BaCo_2As_2 and SrCo_2As_2 exhibit Pauli paramagnetism

down to 2 K³⁹. Moreover, (Ba and Sr)Co₂As₂ have uCT structures whereas CaCo₂As₂ has a cT structure.

Previous reports revealed that electron-doped Sr_{1-x}La_xCo₂As₂ exhibits a sudden change in the magnetic behavior from PM to FM with only 2.5% of La doping^{40,41}, indicating that FM order is preferred over A-type AFM or stripe-type AFM order for this type of doping. A report about the electron doping effects on Sr(Co_{1-x}Ni_x)₂As₂ polycrystalline samples⁴² revealed AFM ordering for $0 < x < 0.3$.

Herein, we report a study of the influence of electron doping in Sr(Co_{1-x}Ni_x)₂As₂ single crystals on the crystallographic, magnetic, thermal, and electronic transport properties. We find that the incorporation of only 1.3% of Ni at the Co sites is sufficient to induce a transition from PM to AFM ground states. This is consistent with our *ab initio* electronic structure calculations which show a peak in the density of states slightly above the Fermi energy due to a flat band. In addition, the ground-state magnetic phase diagram of Sr(Co_{1-x}Ni_x)₂As₂ and the magnetic properties within the frameworks of local-moment magnetism using molecular-field theory and of Takahashi's spin fluctuation theory for weak itinerant ferromagnets are also discussed. The most important result of our studies is the observation of non-Fermi-liquid signatures in the heat capacity and electrical resistivity for compositions in the vicinity of the quantum-critical point at $x \approx 0.30$ between the magnetically-ordered and paramagnetic phases at $T \rightarrow 0$.

The experimental details are given in Sec. II. The crystallographic data and composition analysis are presented in Sec. III. The $M(H)$ and $\chi(T)$ data are presented in Sec. IV and these data are analyzed within a local moment magnetism model in Sec. V. An analysis of the magnetic properties in terms of an itinerant picture is presented in Sec. VI, together with Arrott (M^2 vs H/M) and Takahashi's M^4 vs H/M isotherm plots. The inverse susceptibility in the paramagnetic state above T_N is also discussed within the framework of Takahashi's spin-fluctuation theory. The implications of our electronic structure calculations for the magnetism are also included in this section.

The heat capacity $C_p(T)$ data are presented and discussed in Sec. VII, where non-Fermi-liquid behavior is found at low temperatures. A comparison of the experimental density of states versus x obtained from the $C_p(T)$ measurements with the electronic-structure prediction is also given in this section. Electrical resistivity measurements are presented in Sec. VIII where we discuss non-Fermi-liquid behavior induced by doping and magnetic field. An overall discussion of the non-Fermi-liquid types of behavior in Sr(Co_{1-x}Ni_x)₂As₂ is given in Sec. IX.

A summary of the paper is given in Sec. X. Eight tables of fitted parameters are given in the Appendix.

II. EXPERIMENTAL AND THEORETICAL DETAILS

Single crystals of Sr(Co_{1-x}Ni_x)₂As₂ with compositions ($x = 0, 0.013, 0.04, 0.06, 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9$) were grown using self flux. The starting materials were high-purity elemental Sr(99.999%), Co (99.998%), Ni (99.999%) and As (99.99999%) from Alfa Aesar. A mixture of the elements in a Sr:Co:Ni:As 1:4(1-x):4x:4 molar ratio was placed in an alumina crucible that was sealed under $\approx 1/4$ atm high purity argon in a silica tube. The sealed samples were preheated at 600 °C for 5 h, and then heated to 1300 °C at the rate of 50 °C/h and held there for 15 h. Then the furnace was slowly cooled at the rate of 6 °C/h to 1180 °C. The single crystals were separated from the self flux by decanting the flux with a centrifuge at that temperature. Several 2 to 4 mm size shiny platelike single crystals were obtained from each growth.

Single crystals of SrNi₂As₂ were grown in Bi flux. The sample to flux in a 1:10 molar ratio was placed in an alumina crucible that was sealed under argon gas in a silica tube. A sealed sample was preheated at 600 °C for 6 h. Then the mixture was placed in a box furnace and heated to 1050 °C at a rate of 50 °C/h, held there for 20 h, and then cooled to 700 °C at a rate of 2 °C/h and then to 400 °C at a rate of 5 °C/h. At this temperature the molten Bi flux was decanted using a centrifuge. Shiny platelike crystals with plate-surface dimension 2 to 5 mm (≈ 15 μ m) were obtained.

The phase purity and chemical composition of the Sr(Co_{1-x}Ni_x)₂As₂ crystals were checked by energy dispersive x-ray (EDS) semiquantitative chemical analysis using an EDS attachment to a JEOL scanning-electron microscope (SEM). SEM scans were taken on cleaved surfaces of the crystals which verified the single-phase nature of the crystals. The compositions of each side of a platelike crystal were measured at six or seven positions on each face, which revealed good homogeneity in each crystal. The average compositions and error bars were obtained from these data.

Single-crystal x-ray diffraction (XRD) measurements were performed at room temperature on a Bruker D8 Venture diffractometer operating at tube voltage of 50 kV and a current of 1 mA equipped with a Photon 100 CMOS detector, a flat graphite monochromator and a Mo K α I μ S microfocus source ($\lambda = 0.71073$ Å). The preliminary quality testing has been performed on a set of 32 frames. The raw frame data were collected using the Bruker APEX3 software package⁴³. The frames were integrated with the Bruker SAINT program⁴⁴ using a narrow-frame algorithm integration and the data were corrected for absorption effects using the multi-scan method (SADABS)⁴⁵ within the APEX3 package. The occupancies of the Sr and As sites were set to unity and that of the Co/Ni site was refined. No vacancies on the Co/Ni site were found for any of the crystals. The atomic displacement parameters were refined anisotropically.

ically. Initial models of the crystal structures were first obtained with the program SHELXT-2014⁴⁶ and refined using the program SHELXL-2014⁴⁷ within the APEX3 software package.

The results of the EDS and XRD composition analyses are given in Table I in the Appendix. The same crystals were used to perform the physical-property measurements reported below.

Magnetization data were obtained using a Quantum Design, Inc., magnetic-properties measurement system (MPMS) and a vibrating sample magnetometer (VSM) in a Quantum Design, Inc., physical-properties measurement system (PPMS) in magnetic fields up to 14 T where $1 \text{ T} \equiv 10^4 \text{ Oe}$. Heat capacity $C_p(T)$ measurements were performed in a PPMS system using a relaxation method.

Electrical resistivity measurements were performed with current flow along the tetragonal ab plane. Samples were cleaved in the shape of a bar with typical size $2 \times 0.5 \times 0.1 \text{ mm}^3$. The contacts were made by soldering Ag wires using Sn^{48,49}. The contact resistance was similarly low as in BaFe₂As₂-based superconductors. Four-probe resistivity measurements were performed in a Quantum Design, Inc., PPMS setup. Magnetic fields up to 9 T were applied along the tetragonal c axis.

Density-functional theory^{50,51} (DFT) with local-density approximation^{52,53} (LDA) for exchange-correlation functional were used to solve the KKR-CPA equations using a spherical harmonic basis set within an atomic sphere approximation for scattering sites^{54,55}. A conventional unit cell and $(10 \times 10 \times 4)$ Monkhorst-Pack⁵⁶ k -point mesh was used in the calculations based on the experimental structural parameters.

III. CRYSTAL STRUCTURES

Both SrCo₂As₂ and SrNi₂As₂ adopt the tetragonal ThCr₂Si₂-type crystal structure and the prospective view of its unit cell with selected lattice parameters are shown in Fig. 1. The bonding in the crystal structure has an important impact on the structural and physical properties. The SrCo₂As₂ structure contains a weak As-As interlayer bonding due to the long As-As bond length along the c -axis [$d_{\text{As-As}} = 3.3323(3) \text{ \AA}$] which results in a two-dimensional (2D) Co-As layered structure separated by Sr atoms as shown in Fig. 1(a). Thus SrCo₂As₂ has an uncollapsed-tetragonal structure (ucT). On the other hand, SrNi₂As₂ has a collapsed-tetragonal crystal structure (cT) as it shows a more three-dimensional (3D) Ni-As network via the formation of strong interlayer As-As bonds with $d_{\text{As-As}} = 2.844(11) \text{ \AA}$ along the c axis and with a c/a ratio of 2.466, as listed in Table I. When the As-As interlayer bonding distance approaches the elemental covalent As-As bond distance or when the c/a ratio is less than 2.67, taking CaFe₂As₂ as a reference compound, the system is considered to be in the collapsed-tetragonal phase^{20,57,58}.

Crystallographic data for Sr(Co_{1-x}Ni_x)₂As₂ with

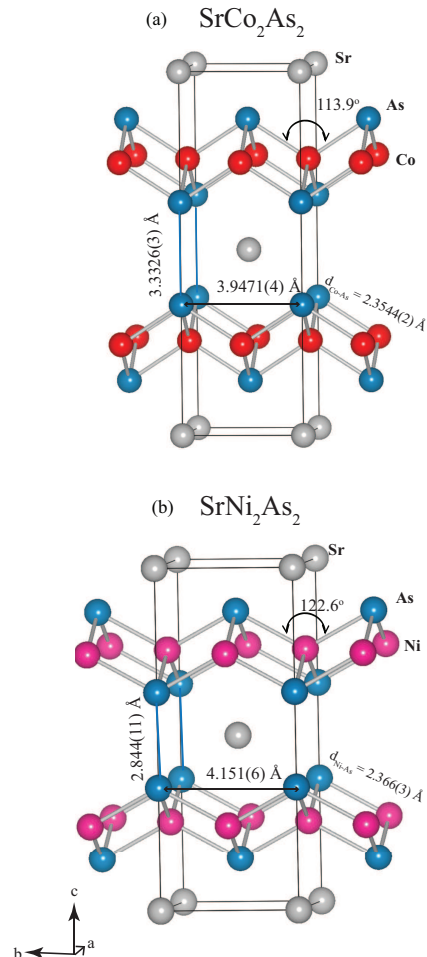


FIG. 1: Unit cell of the crystal structures of uncollapsed-tetragonal (ucT) SrCo₂As₂ and collapsed-tetragonal (cT) SrNi₂As₂ together with selected structural parameters.

$x = 0$ to 1 obtained from the single-crystal XRD measurements at room temperature are given in Table I. The tetragonal lattice parameters a , c , the c -axis As positional parameter z_{As} , the ratio c/a , and the unit cell volume $V_{\text{cell}} = a^2c$ are plotted versus x in Fig. 2. The c lattice parameter strongly but nonlinearly decreases, whereas the a lattice parameter strongly increases approximately linearly with x . These types of behavior give rise to the strongly nonlinear variation of V_{cell} with x . Furthermore, whereas the c/a ratio smoothly and almost linearly decreases with x , the z_{As} varies strongly nonlinearly, suggesting important changes to the electronic properties

The smooth decrease in c/a with x indicates the occurrence of a continuous structural crossover from the ucT structure at $x = 0$ to the cT structure at $x = 1$. The

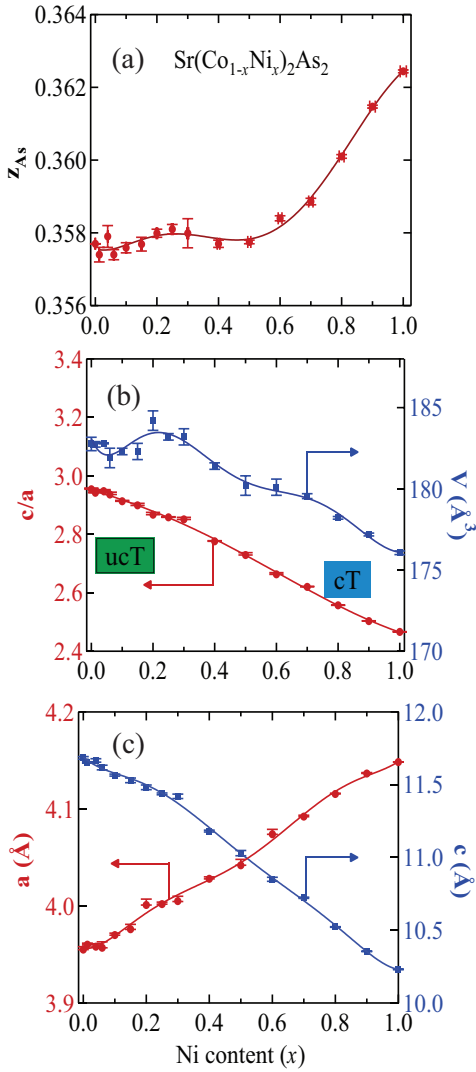


FIG. 2: Crystallographic parameters for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals versus composition x , including (a) z_{As} , the As c -axis position parameter (b) the c/a ratio and unit-cell volume V_{cell} , and (c) the a and c lattice parameters. The solid curves are guides to the eye. A smooth crossover occurs from the uncollapsed-tetragonal (ucT) to collapsed-tetragonal (cT) structures with increasing x .

values of c/a for $x = 0.5$ (2.729) and $x = 0.6$ (2.663) are in the vicinity of the crossover (~ 2.67) between the ucT and cT structures²⁰. Therefore, we infer that a structural crossover occurs in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ at $x \approx 0.5$. It is of interest to study how these structural changes due to electron doping of SrCo_2As_2 correlate with the ground state properties of the system, as investigated in the following sections.

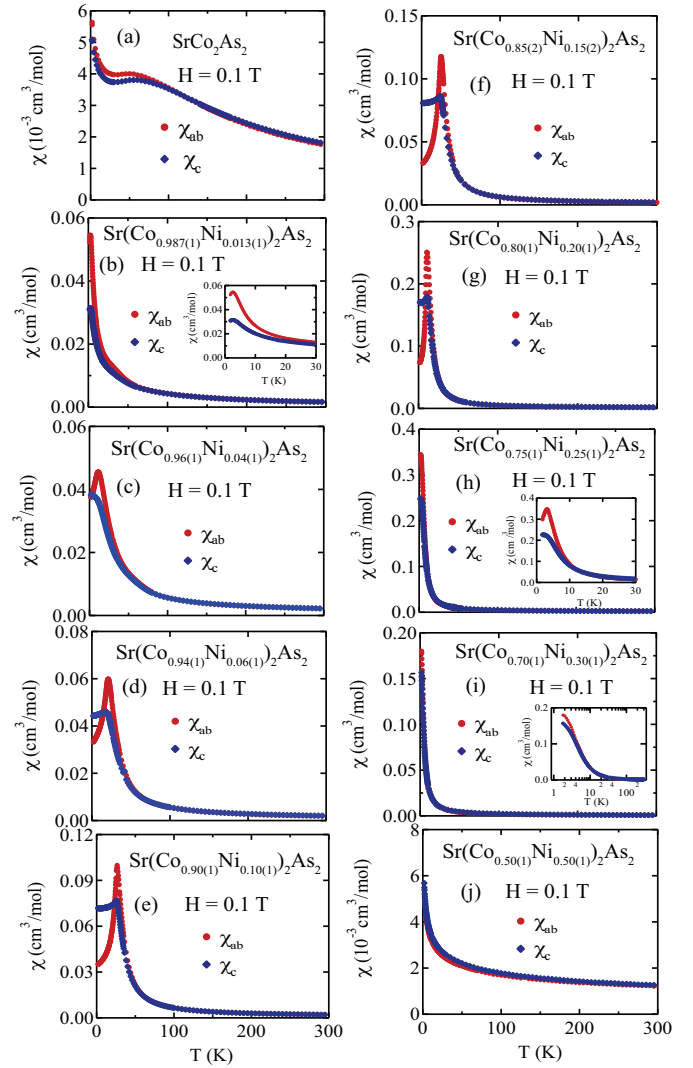


FIG. 3: Zero-field-cooled (ZFC) magnetic susceptibility $\chi \equiv M/H$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0, 0.013, 0.04, 0.06, 0.1, 0.15, 0.20, 0.25, 0.30,$ and 0.50 single crystals as a function of temperature T from 1.8 to 300 K measured in magnetic fields $H = 0.1$ T applied in the ab plane (χ_{ab}) and along the c axis (χ_c).

IV. MAGNETIZATION AND MAGNETIC SUSCEPTIBILITY DATA

The temperature dependence of the magnetic susceptibility $\chi(T) \equiv M(T)/H$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with $x = 0, 0.013, 0.04, 0.06, 0.10, 0.15, 0.20, 0.25, 0.30,$ and 0.50 measured in an applied magnetic field $H = 0.1$ T for both $H \parallel c$ and $H \parallel ab$ are shown in Fig. 3. The self-flux-grown SrCo_2As_2 crystal in Fig. 3(a) has a PM ground state with no observable magnetic ordering transitions down to 0.05 K⁵⁹, supplementing previous data that established this observation for Sn-flux-grown crystals down to 1.8 K²⁸. However, our self-flux-grown crystal shows a significantly larger low- T upturn than Sn-flux-grown crystals do, suggesting a higher level of mag-

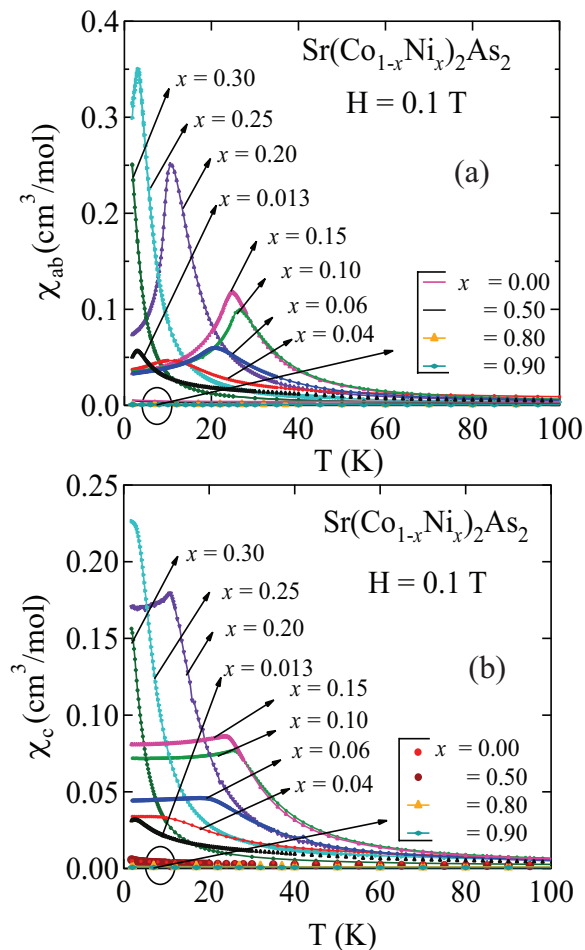


FIG. 4: Comparison of the temperature T dependence of the magnetic susceptibility χ with the applied magnetic field (a) $H \parallel ab$ and (b) $H \parallel c$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with $x = 0$ to 0.90.

netic defects in our crystal. This is consistent with expectation, since our flux was separated from the crystals by centrifuging at 1180 °C, whereas the Sn-flux-grown crystals were centrifuged at the much lower temperature of ~ 700 °C. Probably related to this difference is that the broad maximum in $\chi(T)$ we see at ~ 60 K is significantly lower than the temperature of ≈ 115 K of the broad maximum observed²⁸ for Sn-flux-grown crystals.

A small peak appears in the $\chi(T)$ data in the inset of Fig. 3(b) even for the low doping level $x = 0.013$, ascribed to an AFM transition at $T_N \approx 3$ K. This small peak evolves into larger peaks as the Ni doping increases. The anisotropic $\chi(T)$ data below T_N for $0.04 \leq x \leq 0.20$ in Figs. 3(d)–3(g), where χ_c is nearly independent of T and χ_{ab} strongly decreases for $T \rightarrow 0$, indicate that the AFM ordered moment lies in the ab plane. According to molecular-field theory (MFT), the variable nonzero limits of $\chi_{ab}(T \rightarrow 0)/\chi_c(T_N)$ suggest that the AFM ordering is either an intrinsic coplanar noncollinear ab -plane ordering or intrinsic collinear ab -plane ordering with multiple AFM domains^{60,61}. Comparisons of the ab -plane

and c -axis susceptibilities for the crystals are shown in Figs. 4(a) and 4(b), respectively.

The AFM transition temperatures T_N of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0.013$ to 0.25 were estimated from the temperatures of the maxima of $d(\chi T)/dT$ ⁶² and are listed in Table III. With increasing x , it is seen that T_N initially increases, reaches a maximum of 26.5 K for $x = 0.10$, decreases to 3.1 K for $x = 0.25$, and finally disappears below our low- T limit of 1.8 K for $x \geq 0.30$. The small peak in $\chi(T)$ at $T_N \approx 3$ K for $x = 0.25$ in the inset of Fig. 3(h) is similar to that for $x = 0.013$ in the inset of Fig. 3(b). Thus the ground state of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ changes from PM to AFM with only a small amount of Ni substitution ($x = 0.013$), which shows that the magnetism of SrCo_2As_2 is extremely sensitive to electron doping.

It is also apparent from Figs. 3 and 4 that the $\chi_{ab}(T)$ data near T_N for the crystals with $x = 0.04$ to 0.20 exhibit a strong increase in magnitude compared to crystals with compositions outside this range. This strong increase may due to the occurrence of strong anisotropic FM spin fluctuations beyond MFT as previously found in the $\text{Ca}_{1-x}\text{Sr}_x\text{Co}_{2-y}\text{As}_2$ system³⁶ although single-ion uniaxial anisotropy could also contribute to it. When the ground state of the system changes from AFM to PM for $x \geq 0.50$, the ordinate scale returns approximately to that shown for $x = 0$ in Fig. 3(a).

According to the crystal-structure studies of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ in Sec. III above, the composition $x \sim 0.30$ at which AFM order disappears with increasing x does not coincide with the composition of the crossover between the ucT and cT structures ($x \sim 0.5$). Previous band-structure calculations for SrCo_2As_2 indicated that the system is magnetically metastable due to the presence of a sharp peak in the DOS near E_F ²⁸. We later show in Sec. VID that electron doping increases the $\text{DOS}(E_F)$ due to the presence of a flat band at an energy just above $E_F(x = 0)$.

In order to clarify the nature of the magnetic behavior observed in Figs. 3 and 4 and the variation of the magnetic ground state with Ni doping, magnetization versus applied magnetic field $M(H)$ isotherms were measured at $T = 2$ K for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with compositions $x = 0$ to 0.5 with both $H \parallel ab$ and $H \parallel c$ as shown in Fig. 5. M is proportional to H for $x = 0$, consistent with results from the previous report²⁸. From the $\chi(T)$ data in Fig. 3, the system orders antiferromagnetically already at $x = 0.013$ which is supported by the strongly nonlinear $M(H)$ data in Fig. 5(b). Furthermore, the crystals with $x = 0.04, 0.06, 0.10, 0.15,$ and 0.20 show clear evidence for spin-flop transitions, indicative of ab -plane AFM ordering in agreement with our discussion of the anisotropic $\chi(T)$ above.

Comparisons of the magnetization data for the different crystals are shown for ab -plane and c -axis fields in Fig. 6. One sees that the maximum magnitude of M_{ab} up to $H = 5.5$ T increases upon Ni doping and reaches a maximum at $x = 0.15$, similar to our $\chi(T)$ data in Fig. 4,

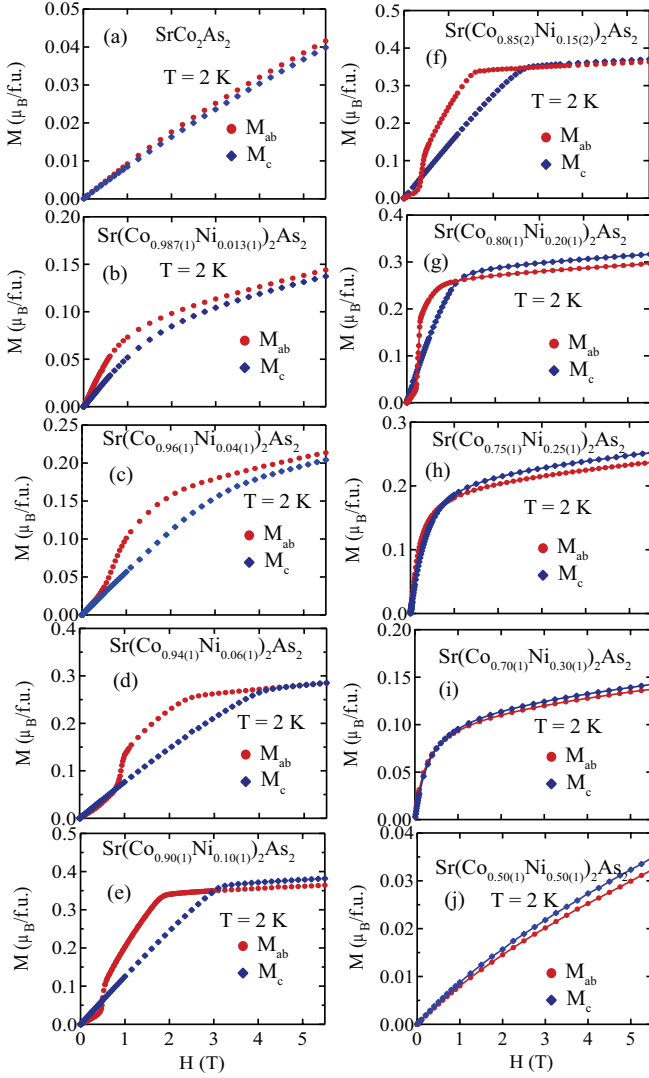


FIG. 5: Magnetization M of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ ($x = 0, 0.013, 0.04, 0.06, 0.1, 0.15, 0.20, 0.25, 0.30,$ and 0.50) single crystals at $T = 2$ K as a function of field H in the range of 0 to 5.5 T applied in the ab plane (M_{ab}) and along the c axis (M_c).

then decreases with further Ni doping. For $x \geq 0.30$, the system is paramagnetic at all temperatures.

The parameters obtained from the $M(H)$ data are the spin-flop field H_{SF} , the critical field H_c at which M_{ab} becomes approximately equal to M_c with increasing H , the saturation magnetic moment M_{sat} obtained from a linear extrapolation of $M(H)$ at $T = 2$ K at high fields to $H = 0$, and the angle-averaged saturation moment $M_{\text{sat,ave}}$ as listed in Table IV and plotted in Figs. 7 and 8. Besides, the spin-flop field H_{SF} first increases from 0.66 T ($x = 0.04$) to 0.91 T ($x = 0.06$), then decreases to 0.23 T ($x = 0.20$) and gradually disappears for $x \geq 0.20$.

In the following sections the $\chi(T)$ and $M(H)$ isotherm data presented above together with additional $M(H)$ isotherm data up to $H = 14$ T are analyzed first from

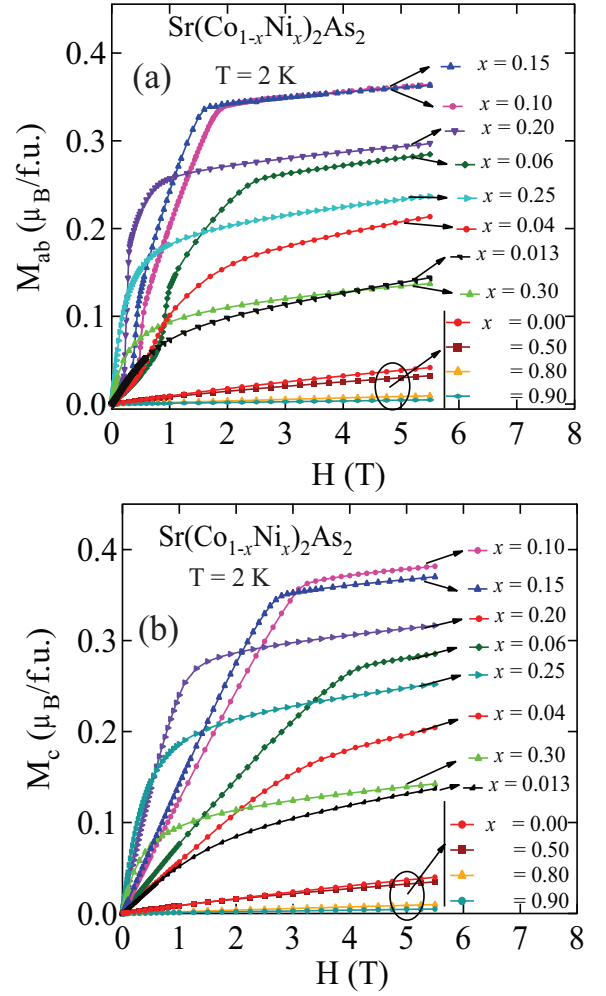


FIG. 6: Comparisons of the applied magnetic-field dependence of the magnetization with fields (a) $H \parallel ab$ and (b) $H \parallel c$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with $x = 0$ to 0.90.

a local-moment perspective and then from an itinerant magnetism point of view.

V. LOCAL-MOMENT MAGNETISM MODEL

A. Curie-Weiss Law for the Paramagnetic State

Here we analyze the magnetic susceptibility $\chi_\alpha(T)$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ at high temperatures (in the PM state) within the local-moment Heisenberg model in terms of the modified Curie-Weiss law

$$\chi_\alpha(T) = \chi_0 + \frac{C_\alpha}{T - \theta_{p\alpha}} \quad (\alpha = ab, c), \quad (1)$$

where χ_0 is a T -independent contribution, C_α is the Curie constant with the field in the α principal-axis direction where α is included to take into account possible magnetic anisotropies, and $\theta_{p\alpha}$ is the Weiss temperature arising from interactions between the spins. For local Heisen-

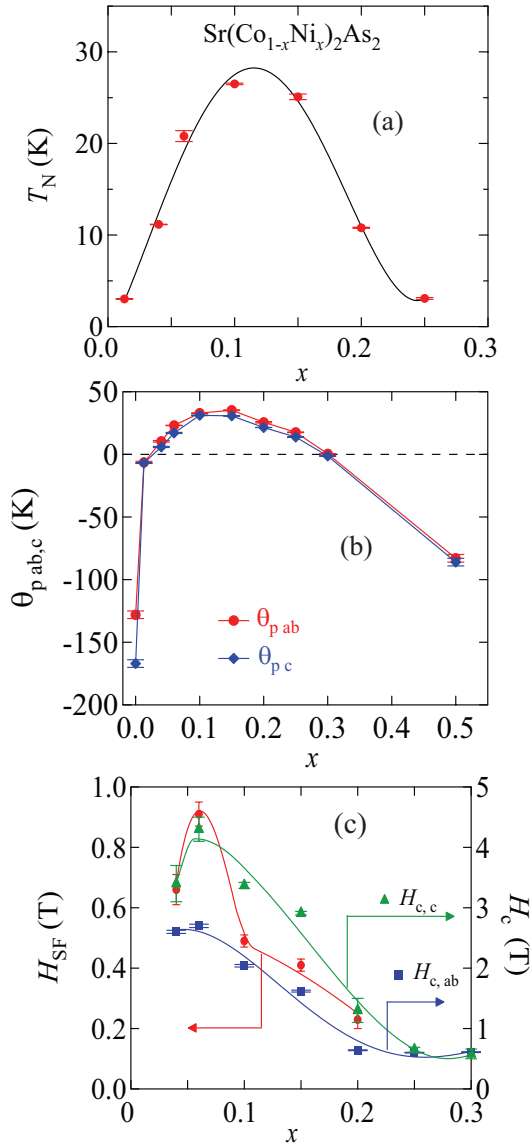


FIG. 7: Composition x dependence of the (a) AFM transition temperature T_N , (b) Weiss temperatures $\theta_{p,ab}$ and $\theta_{p,c}$, and (c) spin-flop field H_{SF} and critical field H_c for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals. One sees that $M_{\text{sat,ave}} \ll \mu_{\text{eff,ave}}$. This indicates that the magnetism is itinerant instead of being due to local magnetic moments. The lines are guides to the eye.

berg spins with $g = 2$ and spin angular momentum quantum number S in units of \hbar , the Curie constant is given in Gaussian cgs units by

$$C \left[\frac{\text{cm}^3 \text{K}}{\text{mol spins}} \right] = 0.5002 S(S+1). \quad (2a)$$

Thus for the minimum quantum spin $S = 1/2$ one obtains

$$C(S = 1/2) = 0.375 \text{ cm}^3 \text{K}/(\text{mol spins}). \quad (2b)$$

Figure 9 shows the inverse susceptibility $\chi^{-1}(T)$ at $H = 0.1$ T applied along the c axis (χ_c^{-1}) and in the

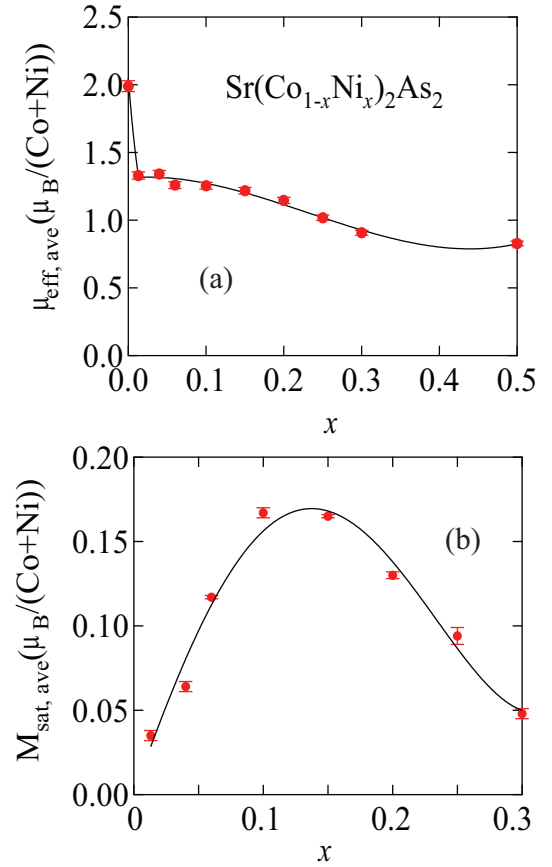


FIG. 8: Composition x dependence of the angle-averaged (a) effective moment per transition metal atom $\mu_{\text{eff,ave}}$ and (b) saturation moment per transition metal atom $M_{\text{sat,ave}}$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals. The lines are guides to the eye.

ab plane (χ_{ab}^{-1}) as a function of T from 1.8 to 300 K. As one can see from the figures, the $\chi^{-1}(T)$ plots are slightly curved, which can be accounted for by a nonzero χ_0 in Eq. (1). The high-temperature ($100 \text{ K} < T < 300 \text{ K}$) $\chi(T)$ data for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0$ to 0.50 for both field directions fitted by Eq. (1) are shown as the solid lines in Fig. 9. The fitting parameters χ_0 , C_α , and $\theta_{p\alpha}$ are listed in Tables II and III together with the effective Bohr magneton number per transition-metal atom $p_{\text{eff}\alpha}$ calculated from C_α using Eq. (4) below. The parameters obtained for SrCo_2As_2 are consistent with the previous single-crystal results²⁸. The temperature-independent susceptibility χ_0 is negative for SrCo_2As_2 but becomes positive upon Ni doping. For $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$, a Co(Ni) atom in the $\text{Co}^{+1}(\text{Ni}^{+2})$ state is expected to have $S = 1$. As a result, from Eq. (2a) one obtains $C = 1 \text{ cm}^3 \text{K}/\text{mol}$. On the other hand, a low spin state $S = \frac{1}{2}$ for $\text{Co}^{+2}(\text{Ni}^{+1})$ gives the Curie constant in Eq. (2b), which is close to the observed values for $0.013 \leq x \leq 0.20$.

The Curie constant C_α is conventionally given per mole

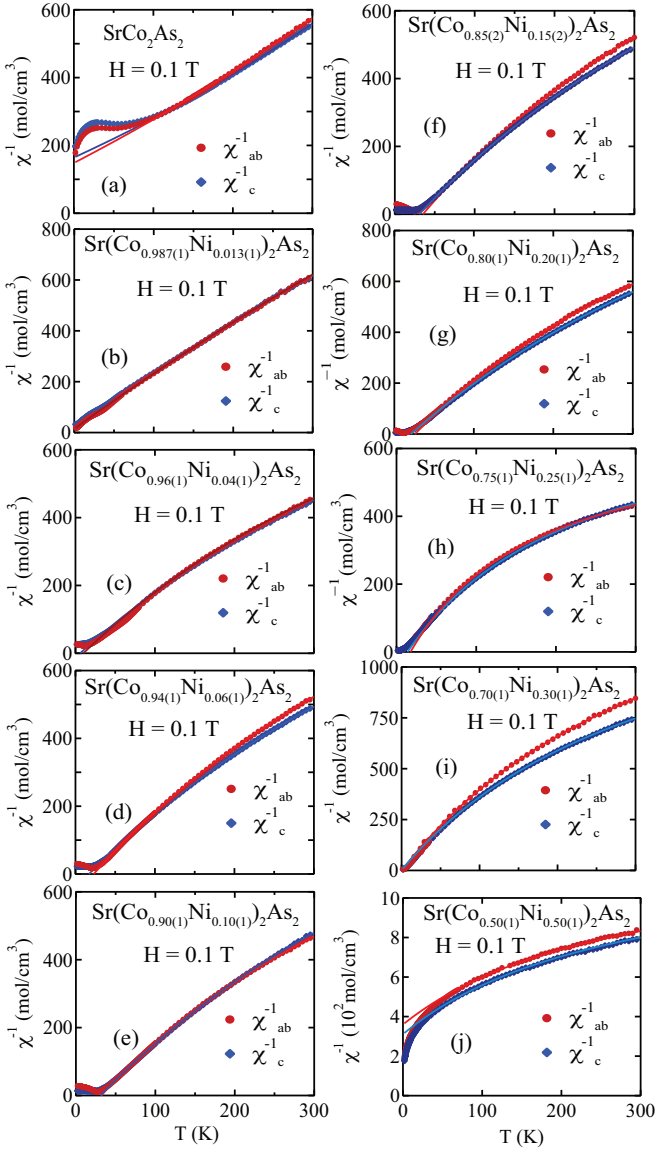


FIG. 9: Zero-field-cooled (ZFC) inverse magnetic susceptibility $\chi^{-1} \equiv H/M$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0, 0.013, 0.04, 0.06, 0.1, 0.15, 0.20, 0.25, 0.30,$ and 0.50 single crystals as a function of temperature T from 1.8 to 300 K measured in magnetic fields $H = 0.1$ T applied in the ab plane (χ_{ab}) and along the c axis (χ_c). The solid curves are fits by Eq. (1).

of formula units by

$$C_\alpha = \frac{N_A g_\alpha^2 S(S+1) \mu_B^2}{3k_B}, \quad (3a)$$

where N_A is Avogadro's number, g is the spectroscopic splitting factor (g factor), S is the spin quantum number of the local moment, μ_B is the Bohr magneton, and k_B is Boltzmann's constant. A convenient parameterization of the Curie constant is in terms of the effective magnetic

moment per formula unit $\mu_{\text{eff}}^{\text{f.u.}}$, given by

$$C_\alpha = \frac{N_A \mu_{\text{eff}\alpha}^{\text{f.u.}2}}{3k_B} = \frac{N_A p_{\text{eff}\alpha}^{\text{f.u.}2} \mu_B^2}{3k_B}, \quad (3b)$$

where $p_{\text{eff}}^{\text{f.u.}}$ is the effective Bohr magneton number per formula unit. If there are n magnetic atoms per f.u., then the effective Bohr magneton number per magnetic atom is given by Eqs. (3a) and (3b) as

$$p_{\text{eff}\alpha} = \sqrt{\frac{3k_B C_\alpha}{n N_A \mu_B^2}} \approx \sqrt{\frac{8C_\alpha}{n}}, \quad (4)$$

where C_α is in cgs units of $\text{cm}^3 \text{K/mol f.u.}$ and the cgs values of the fundamental constants were used. In this paper $n = 2$ for two transition metal atoms per f.u. of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$. Using Eq. (2b), the value of p_{eff} for a local spin-1/2 with $g = 2$ is

$$p_{\text{eff}}(S = 1/2) = 1.73. \quad (5)$$

The values of $p_{\text{eff}\alpha}$ obtained from the respective Curie constants are listed in Tables II and III. With the exception of pure SrCo_2As_2 , the values of $p_{\text{eff}\alpha}$ for our $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals are all significantly smaller than the value in Eq. (5), suggesting itinerant magnetism instead of local-moment magnetism for these compositions.

For later discussion, from Eqs. (3) one has

$$p_{\text{eff}\alpha} = g_\alpha \sqrt{S(S+1)}. \quad (6)$$

Taking a spherical average eliminates the effects of most sources of magnetic anisotropy and one obtains

$$p_{\text{eff}} = g \sqrt{S(S+1)} = \sqrt{gS(gS+g)}. \quad (7)$$

The saturation Bohr magneton p_{sat} (low- T ordered moment divided by μ_B) is

$$p_{\text{sat}} = gS, \quad (8)$$

yielding

$$p_{\text{eff}} = \sqrt{p_{\text{sat}}(p_{\text{sat}} + g)}, \quad (9a)$$

from which one obtains

$$p_{\text{sat}} = \frac{g}{2} \left(\sqrt{1 + \frac{4}{g^2} p_{\text{eff}}^2} - 1 \right). \quad (9b)$$

If one sets $g = 2$ for itinerant carriers and also for Heisenberg spins, one obtains the relation

$$p_{\text{sat}} = \sqrt{1 + p_{\text{eff}}^2} - 1 \quad (g = 2). \quad (9c)$$

In the weak itinerant FM analyses discussed below in Sec. VI, one defines the quantity

$$p_c = \sqrt{1 + p_{\text{eff}}^2} - 1 \quad (g = 2), \quad (9d)$$

where p_{eff} is obtained by fitting the PM susceptibility by the modified Curie-Weiss law (1). Thus p_c is equal to the measured saturation moment p_{sat} for a local-moment system but not generally for an itinerant-electron FM. Hence the ratio p_c/p_{sat} is an indicator of where a particular FM lies in the local-moment/itinerant-moment continuum.

From Table II, SrCo_2As_2 has a large negative Weiss temperature, suggesting that AFM interactions are dominant in the system which is consistent with inelastic neutron scattering measurements where the presence of stripe-type AFM fluctuations was found³⁰. However, the Weiss temperature changes rapidly from $\theta_{p,\text{ave}} = -141$ K ($x = 0$) to $\theta_{p,\text{ave}} = -6.6(2)$ K with a small amount of Ni doping $x = 0.013$, but is still negative. Remarkably, as x increases into the range $0.04 \leq x \leq 0.25$, the Weiss temperature becomes positive (FM-like) and the spherical average increases from $\theta_{p,\text{ave}} = 8.9$ K ($x = 0.04$) to 33.7 K ($x = 0.15$) and then decreases to 16.3 K for $x = 0.25$. The positive Weiss temperatures indicate the presence of predominantly FM exchange interactions, in particular for $x = 0.10$ at which T_N reaches its maximum value. For $x \geq 0.30$, where the system is not magnetically ordered, $\theta_{p,\text{ave}}$ returns to negative values. Paradoxically, $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ thus exhibits the maximum AFM ordering temperatures at compositions at which FM exchange interactions dominate over AFM interactions. The anisotropies in the Curie constants C_α and in the Weiss temperatures $\theta_{p\alpha}$ in Table II may arise from g anisotropy, single-ion uniaxial anisotropy, anisotropic magnetic dipole interactions, and/or from exchange anisotropy.

B. Molecular-Field-Theory Analysis of Magnetic Susceptibility in the Ordered State in Terms of Helical AFM Ordering

In this section we use the so-called unified molecular field theory (MFT) for local-moment AFMs developed by one of the authors^{60,61}. This theory applies to systems of identical crystallographically-equivalent Heisenberg spins interacting by Heisenberg exchange and does not use the concept of magnetic sublattices. Instead, the zero-field magnetic properties are calculated solely from the Heisenberg exchange interactions between an arbitrary spin and its neighbors. This formulation will be used to calculate the anisotropic $\chi(T)$ at temperatures $T \leq T_N$ arising from planar helical AFM ordering with the ordered moments aligned in the ab plane where the helix axis is the c axis.

A picture of a z -axis helix is shown in Fig. 10, where here the xy plane corresponds to the ab plane and the z axis to the c axis. Within the classical J_0 - J_1 - J_2 local-moment mean-field Heisenberg model described in the figure caption, the turn angle kd between adjacent FM-

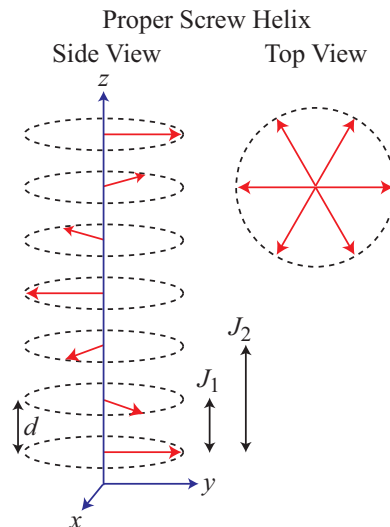


FIG. 10: Generic helical AFM structure⁶⁰. Each arrow represents a layer of moments perpendicular to the z axis that are ferromagnetically aligned within the xy plane and with inter-layer separation d . The wave vector \mathbf{k} of the helix is directed along the z axis. The magnetic moment turn angle between adjacent magnetic layers is kd . The nearest-layer and next-nearest-layer exchange interactions J_1 and J_2 , respectively, within the J_0 - J_1 - J_2 Heisenberg MFT model are indicated. The top view is a hodograph of the magnetic moments.

aligned planes is given by^{60,61}

$$kd = \arccos\left(-\frac{J_1}{4J_2}\right), \quad (10)$$

where J_2 must be positive (AFM) for a helical structure, whereas J_1 can be either negative (FM) or positive. Within MFT, the c -axis susceptibility χ_{Jc} of the helix is independent of temperature for $T \leq T_N$, whereas the ratio of the in-plane zero-field susceptibility χ_{Jab} at $T = 0$ to $\chi_J(T_N)$ is^{60,61}

$$\frac{\chi_{Jab}(T = 0)}{\chi_J(T_N)} = \frac{1}{2[1 + 2 \cos(kd) + 2 \cos^2(kd)]}, \quad (11)$$

and the subscript J indicates that anisotropy and T -independent terms in $\chi(T \geq T_N)$ are not present in the data or have been corrected for.

In order to compare our $\chi(T \leq T_N)$ data in Fig. 3 with Eq. (11), we first correct for the T -independent susceptibility χ_0 and the anisotropy at $T > T_N$. χ_0 is taken into account at all temperatures according to

$$\chi_{J\alpha}(T) = \chi_\alpha(T) - \chi_{0\alpha} \quad (\alpha = ab \text{ or } c), \quad (12a)$$

where $\chi_\alpha(T)$ is the measured susceptibility and the $\chi_{0\alpha}$ values are given in Table II. The remaining anisotropy in the PM state is likely due to magnetic dipole interactions and/or single-ion quantum uniaxial anisotropies, for which the magnetic susceptibility tensor is traceless

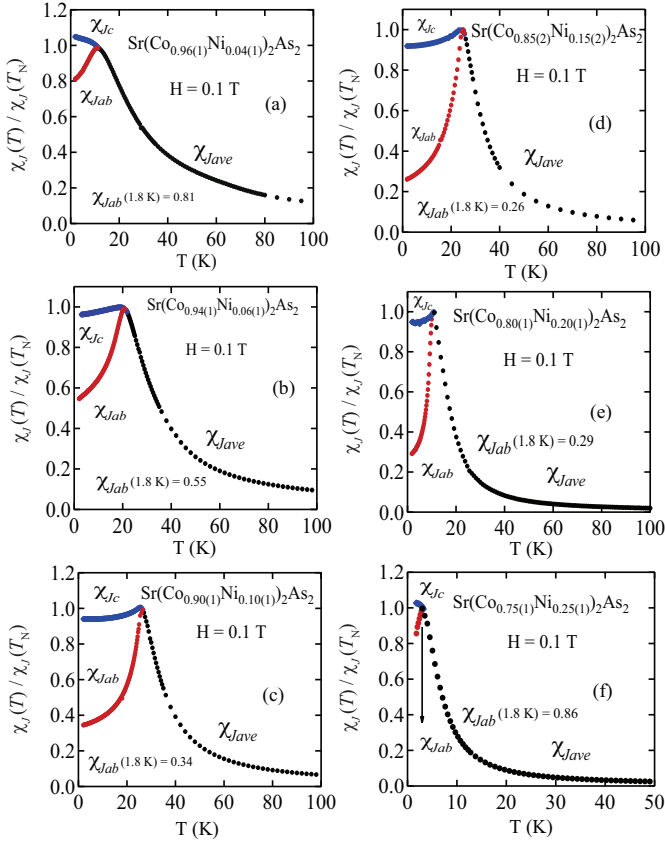


FIG. 11: Normalized susceptibility $\chi_J(T)/\chi_J(T_N)$ versus T of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ for $x = 0.04, 0.06, 0.1, 0.15, 0.20$, and 0.25 at applied field $H = 0.1$ T in the ab plane χ_{Jab} and along the c axis χ_{Jc} .

in the PM state. Then one obtains the Heisenberg susceptibility χ_J in the PM state according to

$$\chi_J(T \geq T_N) = \frac{1}{3}[2\chi_{ab}(T) + \chi_c(T)] \equiv \chi_{Jave}(T \geq T_N). \quad (12b)$$

Figure 11 shows the normalized $\chi_{Jab}(T)/\chi_J(T_N)$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0.04, 0.06, 0.10, 0.15, 0.20$, and 0.25 after correcting for χ_0 and the anisotropy in the PM state as discussed above. One can now estimate the helix turn angle kd from $\chi_J(T = 1.8 \text{ K})/\chi_J(T_N)$ using Eq. (11) and the results are listed in Table III and plotted in Fig. 12, where kd is seen to depend approximately quadratically on the Ni content x . The magnetic structure may be incommensurate because the values of kd appear to be irrational.

The above MFT discussion of helical ordering may be expected to be of only qualitative significance, since the MFT was formulated for local-moment AFMs instead of itinerant AFMs. However, magnetic neutron diffraction measurements^{63,64} found that the magnetic structure is indeed helical, with helix turn angles that decrease from 104° to 94° on increasing x from $x = 0.06$ to 0.15 , which have the same trend and are of the same order as we infer in Fig. 12 from Eq. (11) for local-moment helical

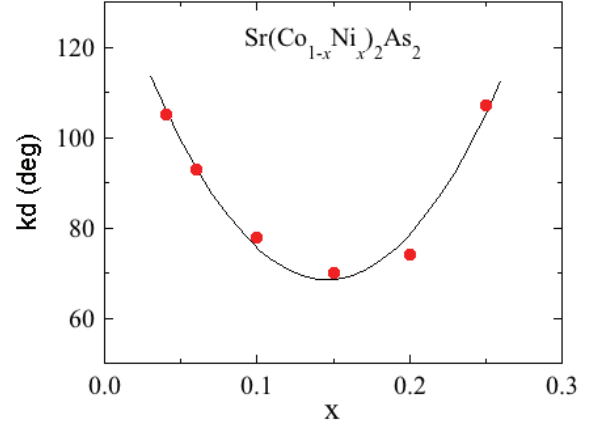


FIG. 12: Turn angle kd versus composition x for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals inferred from Fig. 11 using Eq. (11) for local-moment helical Heisenberg antiferromagnets. The solid black curve is a quadratic fit to the data.

Heisenberg AFMs.

VI. ITINERANT FERROMAGNETISM MODEL

The $\chi^{-1}(T)$ data in Fig. 9 for our $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals follow Curie-Weiss-like behavior in the PM state with $T > T_N$ with p_{eff} values as given in Table II which are smaller than expected for the minimum local spin $S = 1/2$ in Eq. (5), suggestive of itinerant moments. Furthermore, the small angle-averaged saturation moments $\mu_{\text{sat,ave}} \sim 0.1 \mu_B$ per transition metal atom in Table IV also suggest itinerant magnetism. Thus the magnetism in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ is likely itinerant rather than arising from local moments. In addition, the Weiss temperatures for the crystals with the larger T_N values are positive, indicating dominant FM interactions. Therefore in this section we analyze the high-field $M(H)$ data and the $\chi(T)$ data in the PM regime for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ using Takahashi's spin-fluctuation theory of weak (small ordered moment) itinerant ferromagnets⁶⁵.

A. M^2 vs H/M Isotherms, M^4 vs H/M Isotherms, and the Curie Temperature for Weak Itinerant Ferromagnetism in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$

Figure 13 displays $M(H)$ isotherms with H up to 14 T for six $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals at the indicated temperatures. It is seen that the saturation moment obtained from an extrapolation of the high-field data to $H = 0$ is very small, of order $0.1 \mu_B/(\text{Co}+\text{Ni})$, and that the $M(H)$ curves at $T = 2$ K continue to increase with increasing magnetic field and do not saturate up to 14 T, as expected for an itinerant magnetic system. We find that our data can be well understood in terms of Takahashi's theory of spin-fluctuation-driven weak itinerant

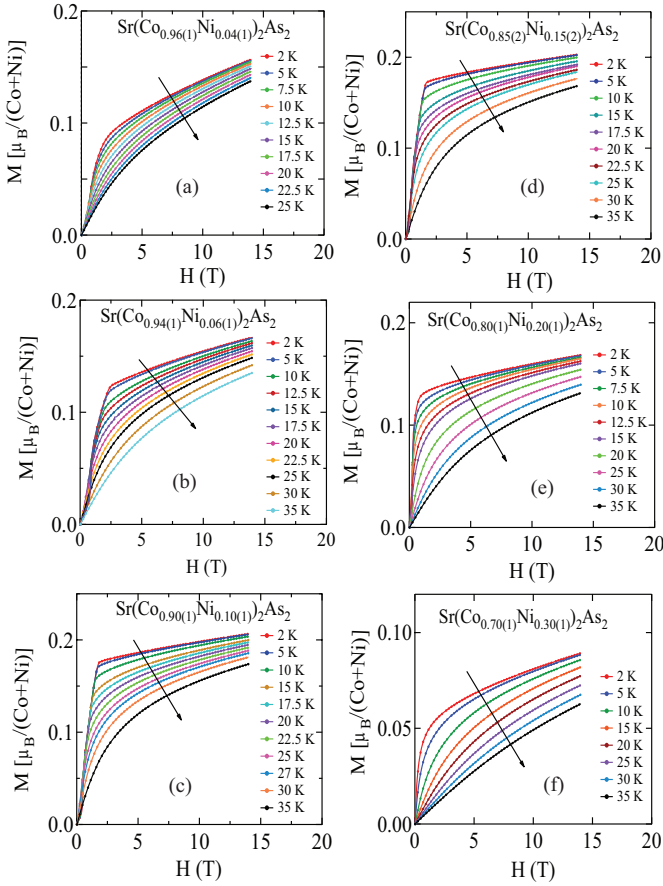


FIG. 13: Isothermal magnetization M vs H of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with $x = 0.04, 0.06, 1, 0.15, 0.20,$ and 0.3 as a function of magnetic field H (0 to 14 T) applied in the ab plane at the indicated temperatures.

ferromagnetism⁶⁵, as follows.

In Takahashi's theory, it is assumed that the total local spin fluctuation amplitude, i.e., the sum of zero point and thermally-induced spin fluctuation amplitudes, is globally conserved (independent of temperature) for weak ferromagnets. The phenomenological Landau expansion of the free energy functional $F(M, T)$ for a ferromagnet in terms of the magnetization per unit volume M at temperature T up to sixth order in M is

$$F(M, T) = F(0, T) + \frac{1}{2}a_2(T)M^2 + \frac{1}{4}a_4(T)M^4 + \frac{1}{6}a_6(T)M^6 - MH \quad (13)$$

where the a_n are expansion coefficients that depend on T in general and the magnetization \mathbf{M} is in the direction of the applied magnetic field \mathbf{H} . Only even powers of M occur because the sign of F cannot depend on the direction of the applied field. Minimizing the free energy (13) with respect to the magnetization $[\partial F(M, T)/\partial M = 0]$ gives the generic thermal-equilibrium prediction

$$H = a_2(T)M + a_4(T)M^3 + a_6(T)M^5. \quad (14)$$

For the ground state ($T = 0$), Takahashi obtained⁶⁵

$$H = \frac{F_1}{(g\mu_B)^4 N_0^3} (M^2 - M_0^2)M, \quad (15)$$

$$F_1 = \frac{2k_B T_A^2}{15cT_0},$$

where a_6 in Eq. (14) is zero, $c = 1/2$ and N_0 is the number of magnetic atoms. One sees that in the ground state, the magnetic isotherm is influenced by the presence of zero-point spin fluctuations because F_1 is expressed in terms of the ferromagnetic spin fluctuation parameters T_0 and T_A in temperature units (K). The parameter T_A is a measure of the width of the distribution of the dynamical spin fluctuations in wave vector space, whereas T_0 represents the energy width of the spin fluctuation spectrum in energy (frequency) space. These two parameters can be experimentally from the measured Arrott plots (M^2 versus H/M isotherms⁶⁶) at low temperatures as follows.

Utilizing the values of k_B and μ_B in cgs units with $g = 2$, Eq. (15) gives

$$H(\text{Oe}) = \left(248.2 \frac{\text{Oe}}{\text{K}}\right) \left(\frac{T_A^2}{T_0}\right) \frac{(M^2 - M_0^2)M}{(N_0\mu_B)^3}. \quad (16)$$

Now expressing M and M_0 in units of μ_B per magnetic atom gives

$$H(\text{Oe}) = \left(248.2 \frac{\text{Oe}}{\text{K}}\right) \left(\frac{T_A^2}{T_0}\right) (M^2 - M_0^2)M, \quad (17)$$

which yields the Arrott-like expression

$$M^2 - M_0^2 = \frac{1}{(248.2 \frac{\text{Oe}}{\text{K}}) \left(\frac{T_A^2}{T_0}\right)} \frac{H}{M}, \quad (18a)$$

where M and M_0 are now dimensionless because their units have been divided out and H is in cgs units of Oe. Thus a plot of M^2 vs H/M is predicted to be linear with y -intercept M_0 and slope

$$\frac{M^2 - M_0^2}{H/M} = \frac{1}{(248.2 \frac{\text{Oe}}{\text{K}}) \left(\frac{T_A^2}{T_0}\right)}. \quad (18b)$$

The ordered moment M_0 in the ground state, again in dimensionless units of μ_B per magnetic atom, is given by⁶⁵

$$M_0 \approx 2\sqrt{C_{4/3} \frac{5T_0}{T_A}} \left(\frac{T_C}{T_A}\right)^{2/3}, \quad (18c)$$

where $C_{4/3}$ is a constant ($C_{4/3} = 1.006089\dots$).

Equations (18b) and (18c) allow both spectral parameters T_A and T_0 to be estimated from the ground-state $M(H)$ isotherm, provided a value for the inferred Curie temperature T_C is known. Here one expects Eqs. (18) for the ground-state magnetization isotherm to be valid for a particular ferromagnet if the minimum measurement

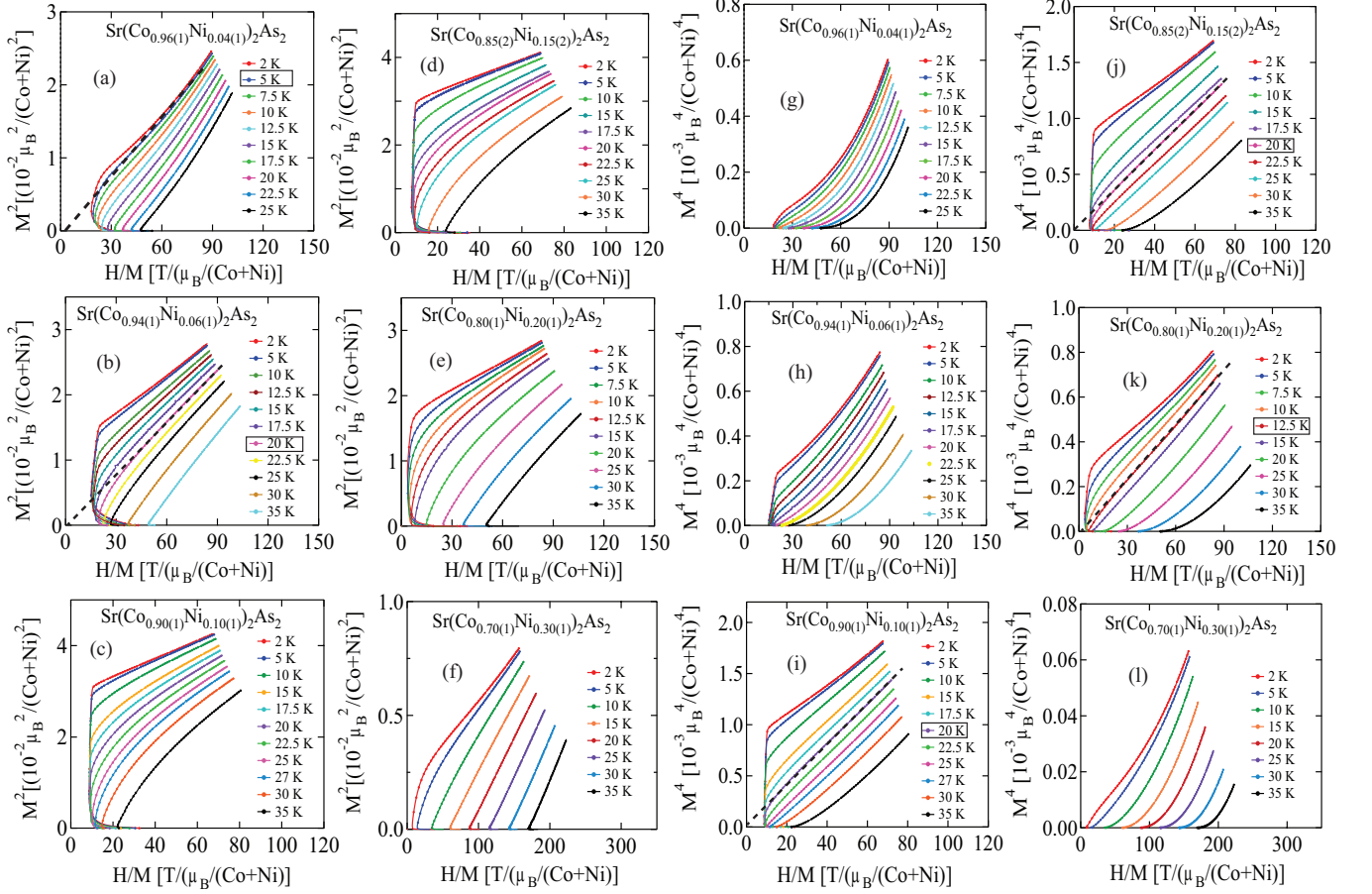


FIG. 14: (a)–(f): Isothermal magnetization M^2 vs H/M plots (Arrott plots) of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with compositions $x = 0.04, 0.06, 1, 0.15, 0.20,$ and 0.3 as a function of magnetic field H applied in the ab plane at the indicated temperatures. Only the higher-field data are relevant due to the presence of spin-flop transitions at lower fields. The temperatures of the isotherms for which dashed straight line extrapolations to low field pass through the origin (a) and (b) with $x = 0.04$ and 0.06 , respectively, are inferred to be the Curie temperatures T_C for the respective samples. (g)–(l): Isothermal magnetization M^4 vs H/M for the same $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals as a function of magnetic field H applied in the ab plane at the indicated temperatures.

temperature for the high-field $M(H)$ isotherms is much less than the measured Curie temperature T_C .

The above low-temperature ground-state isotherm of M^2 versus H/M is predicted to be linear at high fields. In contrast, the critical isotherm at T_C has the different form M^4 versus H/M which shows a proportional behavior with zero y intercept⁶⁵. This form arises because the critical magnetic isotherm is predominantly influenced by temperature-induced spin fluctuations instead of zero-point spin fluctuations.

The critical isotherm at T_C is described by⁶⁵

$$H = \frac{T_A^3 k_B / \mu_B}{2[3\pi(2 + \sqrt{5})T_C]^2} \left(\frac{M}{N_0 \mu_B} \right)^5, \quad (19)$$

$$= \left(4.671 \frac{\text{Oe}}{\text{K}} \right) \left(\frac{T_A^3}{T_C^2} \right) \left(\frac{M}{N_0 \mu_B} \right)^5,$$

where here both a_2 and a_4 in Eq. (14) are zero. Again expressing M in units of μ_B per magnetic atom makes

M dimensionless and gives the proportional relation in Gaussian cgs units as

$$M^4 = \frac{1}{4.671 \frac{\text{Oe}}{\text{K}} \left(\frac{T_A^3}{T_C^2} \right)} \frac{H}{M} \quad (20)$$

with slope

$$\frac{M^4}{H/M} = \frac{1}{4.671 \frac{\text{Oe}}{\text{K}} \left(\frac{T_A^3}{T_C^2} \right)}. \quad (21)$$

Thus at T_C , M^4 is proportional to H/M with no y intercept. The slope is expressed in terms of T_A , which allows T_A to be determined since T_C can be determined from the temperature at which an extrapolation of the high-field linear behavior passes through the origin on an M^4 versus H/M plot.

We analyze our ab -plane magnetization data at fields up to $H = 14$ T for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with compositions $x = 0.04, 0.06, 0.10, 0.15, 0.20,$ and 0.30 in

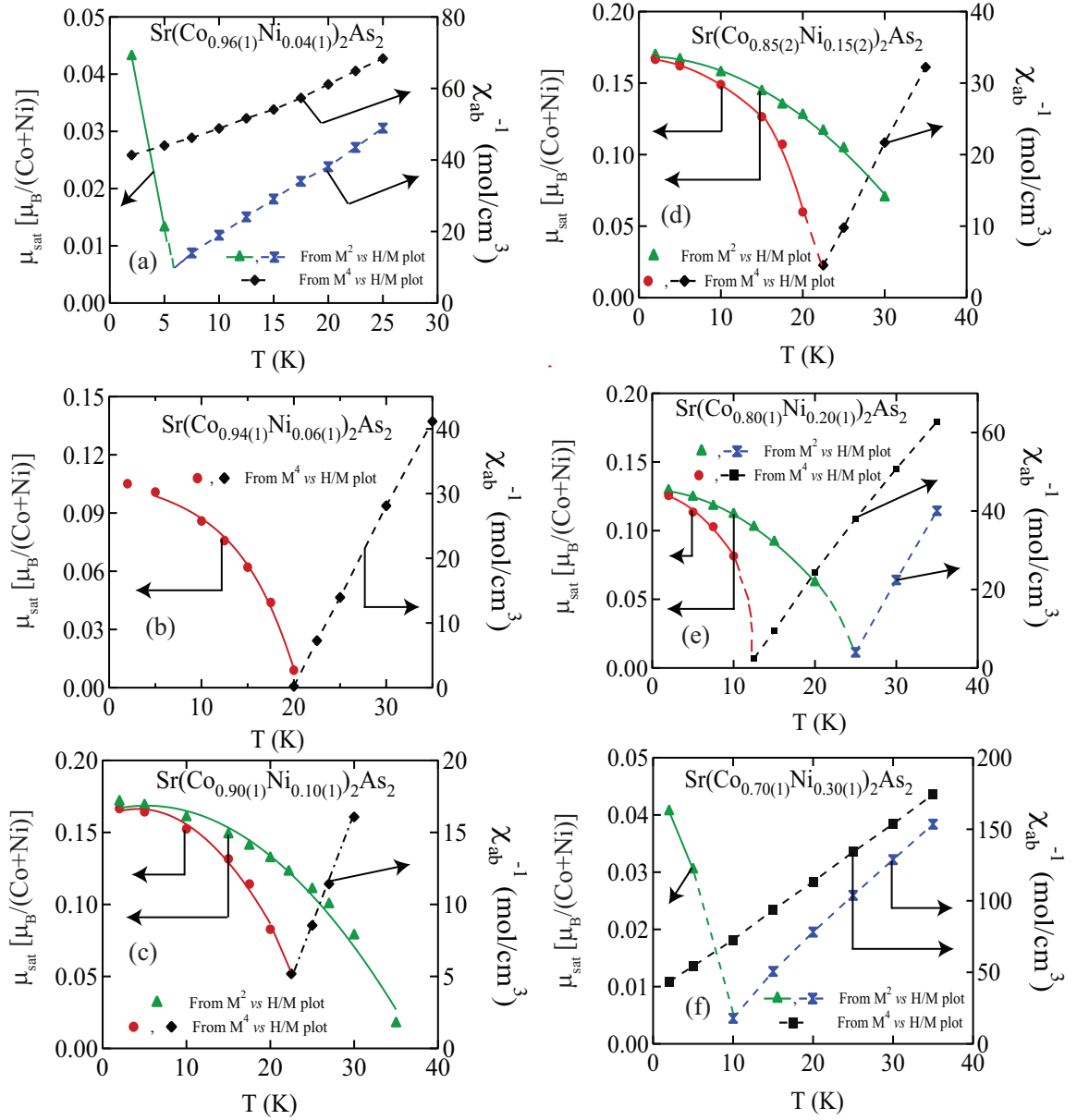


FIG. 15: Saturation moment μ_{sat} and inverse molar susceptibility χ_{ab}^{-1} extracted from the M^2 vs H/M and M^4 vs H/M measurements in Fig. 14 plotted with left and right ordinate scales, respectively.

Fig. 14 in terms of M^2 vs H/M (Arrott) and M^4 versus H/M plots, respectively. The data at the higher magnetic fields was used for the analyses since spin-flop transitions occur at lower fields (see Table IV). The temperatures at which the extrapolations of the high-field data to $H = 0$ pass through the origin are indicated by solid dashed lines in Figs. 14(i), 14(j), and 14(k) for $x = 0.10$, 0.15, and 0.20, respectively. The corresponding values of the saturation moment μ_{sat} and the inverse susceptibility χ_{ab}^{-1} obtained from the extrapolated intercepts of the linear fits to the y and x axes, respectively, are shown in Fig. 15. Here, the temperature at which the extrapolated $\chi_{\text{ab}}^{-1}(T)$ passes through zero determines the Curie temperature T_C .

As one can see in Fig. 14, Arrott plots are only relevant for $x = 0.04$, 0.06, and 0.30 since they show reasonably linear variations at high fields. At the same time, the Arrott plots for $x = 0.10$, 0.15, and 0.20 show strong curvature around T_C . In this case, one needs to analyze the isothermal $M(H)$ data in terms of M^4 vs H/M plots, shown in Fig. 14 (g)–(l), where these plots for $x = 0.10$, 0.15, and 0.20 show linear variations at high fields. Conversely, the M^4 vs H/M plots for $x = 0.04$, 0.06, and 0.30 show an upward curvature for temperatures around T_C . The values of T_C for which the extrapolated data pass through the origin are thus determined from the Arrott plots for $x = 0.04$, 0.06 and from the M^4 versus H/M plots for $x = 0.10$, 0.15, and 0.20 and are listed

in Table V. These differences indicate that temperature-induced spin fluctuations are not as important for the compositions $x = 0.04$ and 0.06 as for $x = 0.10, 0.15,$ and 0.20 . In this context, it is interesting that the critical isotherm at $T = 30$ K for the itinerant ferromagnet $\text{SrCo}_2(\text{Ge}_{1-x}\text{P}_x)_2$ with $x = 0.6$ was linear on an Arrott plot³⁸, which is analogous to our crystals with $x = 0.04$ and 0.06 .

The spin fluctuation parameters T_A and T_0 are calculated based on Arrott plots using Eqs. (18b) and (18c), whereas Eqs. (18c) and (21) are used to calculate T_A and T_0 from the M^4 vs H/M isotherms at T_C and all the resulting derived parameters are listed at the top and bottom of Table V, respectively. The parameter F_1 is calculated using Eq. (15).

B. Rhodes-Wohlfarth Plots

For weak itinerant ferromagnets, the spontaneous (ordered) magnetic moment is much smaller than expected for local-moment magnets, *i.e.*, significantly smaller than minimum value of $1 \mu_B$ per magnetic atom for $S = 1/2$ and $g = 2$. The effective moment can also be smaller than the minimum value of $1.73 \mu_B$ per magnetic atom for $S = 1/2$ in Eq. (5). To systematize these types of behavior, Rhodes and Wohlfarth proposed to plot the ratio p_c/p_{sat} versus the Curie temperature T_C ⁶⁷, now called Rhodes-Wohlfarth plot, where p_c is defined above in Eq. (9d) in terms of the measured effective moment μ_{eff} obtained at $T > T_C$ from the Curie-Weiss law describing the magnetic susceptibility in the PM phase.

As previously noted in Sec. V A, the ratio $p_c/p_{\text{sat}} = 1$ for local-moment magnets, whereas for itinerant-moment systems, one obtains $p_c/p_{\text{sat}} > 1$. Listed in Table VI are the Rhodes-Wohlfarth ratios p_c/p_{sat} calculated for our $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with compositions $x = 0.04, 0.06, 0.10, 0.15, 0.20, 0.25,$ and 0.30 from the data in Tables III and V. Figure 16(a) shows a plot of p_c/p_{sat} versus both T_C and the angle-averaged Weiss temperature $\theta_{p,\text{ave}}$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$. One sees that $p_c/p_{\text{sat}} > 1$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ as expected. For $0.10 \leq x \leq 0.25$ where $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ orders magnetically, the value of p_c/p_{sat} is close to that for the famous weak itinerant ferromagnet ZrZn_2 ⁶⁸. For $x = 0.30$, the ratio increases to 6.1.

Another generalized plot of $p_{\text{eff}}/p_{\text{sat}}$ vs T_C/T_0 , known as the Deguchi-Takahashi plot, is also based on the spin-fluctuation theory. The ratio T_C/T_0 characterizes the degree of itinerancy of the magnetic electrons in the spin fluctuation theory. The value of T_C/T_0 becomes small for weak itinerant ferromagnets with a large temperature-induced spin fluctuation amplitude. A value $T_C/T_0 \sim 1$ is the localized-moment limit. The obtained values of T_C/T_0 estimated from the M^2 vs H/M and M^4 vs H/M isotherms are listed in Table VI and a plot of $p_{\text{eff}}/p_{\text{sat}}$ versus T_C/T_0 is shown in Fig. 16(b). These

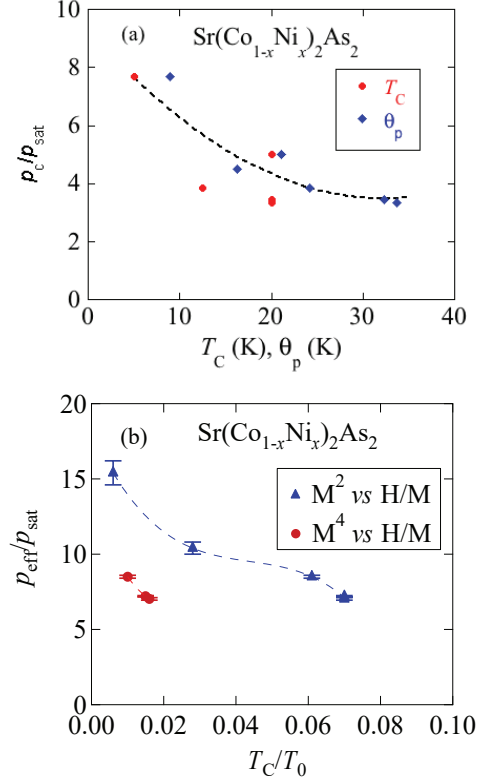


FIG. 16: (a) Rhodes-Wohlfarth plot p_c/p_{sat} vs Curie temperature T_C and average Weiss temperature $\theta_{p,\text{ave}}$. (b) Deguchi-Takahashi plot of $p_{\text{eff}}/p_{\text{sat}}$ vs T_C/T_0 for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with $x = 0.04, 0.06, 0.10, 0.15, 0.20,$ and 0.25 . The dashed curves are guides to the eye.

data indicate that the magnetically-ordered samples of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ are close to the itinerant-magnetism limit of the itinerant magnetism-local moment magnetism continuum.

C. Magnetic Susceptibility of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ above T_C in Terms of Takahashi's Spin-Fluctuation Theory for Weak Itinerant Ferromagnets

The dimensionless parameter y in Takahashi's theory of the thermal-fluctuation-induced magnetic susceptibility χ above T_C is given by⁶⁵

$$y = \frac{N_A g^2 \mu_B^2}{2k_B T_A \chi}, \quad (22a)$$

where N_A is Avogadro's number, $g = 2$, and χ is in cgs units of $\text{cm}^3/(\text{mol magnetic atoms})$. From Eq. (22a) one obtains the inverse magnetic susceptibility χ^{-1} as

$$\chi^{-1} = \left(\frac{2k_B T_A}{N_A g^2 \mu_B^2} \right) y. \quad (22b)$$

The temperature-induced amplitude $A(y, t)$ of the spin

fluctuations is given by⁶⁵

$$A(y, t) = \int_0^1 \left\{ \ln[u(x)] - \frac{1}{2u(x)} - \psi[u(x)] \right\} x^3 dx, \quad (22c)$$

where

$$u(x) \equiv x(x^2 + y)/t, \quad t = T/T_0, \quad (22d)$$

and $\psi(z)$ is the digamma function which is the logarithmic derivative of the gamma function $\Gamma(z)$. The parameter y in Eq. (22b) is numerically calculated from⁶⁵

$$y(t) = \frac{1}{c} [A(y, t) - A(0, t_C)], \quad (22e)$$

where $t_C \equiv T_C/T_0$ and $c = 1/2$.

Since the experimental data are normalized to a mole of formula units of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ where a f.u. contains two magnetic atoms, whereas the theoretical prediction is normalized to a mole of magnetic atoms, the theoretical prediction to compare with experiment is given by

$$\frac{2}{\chi(\text{cm}^3/\text{mol magnetic atoms})} = \frac{1}{\chi(\text{cm}^3/\text{mol f.u.})}, \quad (23)$$

where the theoretical χ^{-1} on the left-hand side is given in Eq. (22b). The black dashed lines and the solid green lines in Fig. 17 are the results obtained using the spin fluctuation parameters T_{A1} , T_{A2} and T_{02} given in Table V, whereas the solid blue lines are fits of the respective experimental data by Eqs. (22) using T_A , labeled as T_{A^*} , as an adjustable parameter. The values of T_C utilized in the fits are also given in Table V. The calculated inverse susceptibility is seen to follow a Curie-Weiss-type law rather well for $T > T_C$ for all the theoretical plots, and is seen to sensitively depend on the value of T_A .

D. Electronic Structure Calculations: Magnetism

To explain the observed magnetic ordering in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ from an itinerant magnetism picture, we calculated the spectral functions (electronic dispersion, or band structure) of the alloyed compounds via the KKR-CPA method^{69–71}, which handles chemical and magnetic disorder^{54,55,72–74}. For pure SrCo_2As_2 [$x = 0$, Fig. 18(a)], segments of flat bands along the $\Gamma - X - Y$ and $P - Y_1 - Z$ directions (or $M - \Gamma$ and $A - Z$ directions in the conventional cell) are just 100 meV above the Fermi energy E_F . These flat bands are derived mostly from Co $d_{x^2-y^2}$ orbitals. The irreducible Brillouin zone of the body-centered tetragonal primitive unit cell of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ is shown in Fig. 19 and is labeled with high-symmetry points and lines that are used in Fig. 18.

Substituting Ni on the Co sites corresponds to electron doping. For $x = 0.15$, the flat bands along $\Gamma - X$ and $P - Y_1 - Z$ directions [Fig. 18(b)] already touch E_F , which can drive Fermi-surface nesting and Stoner-type instabilities.

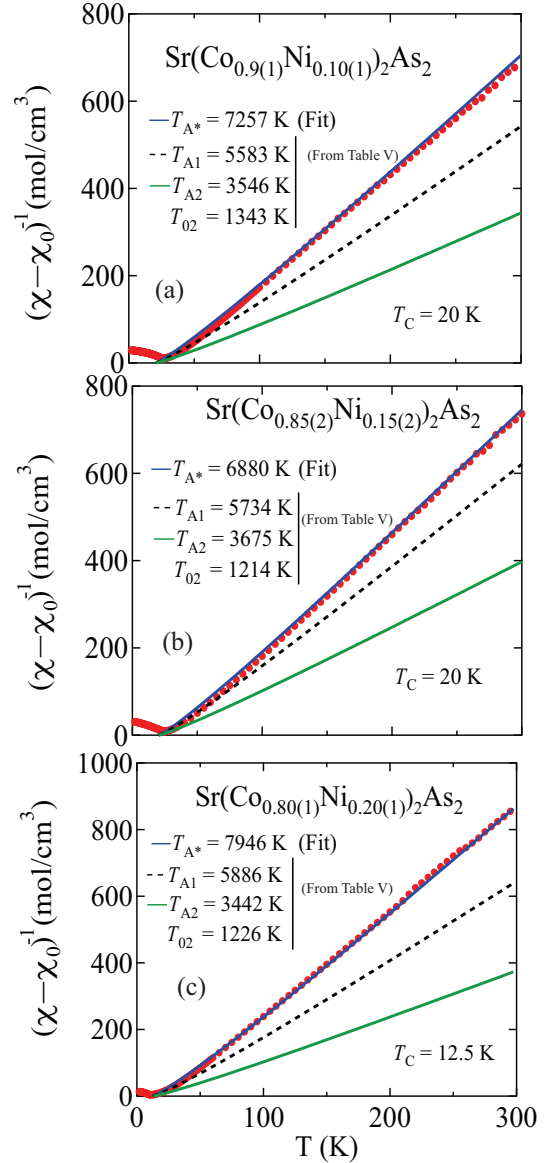


FIG. 17: Temperature dependence of the inverse susceptibility $(\chi - \chi_0)^{-1}$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0.10, 0.15$, and 0.20 . The red circles represent experimental data. The black dashed lines and the green lines are data calculated using the T_A , T_0 , and T_C values listed in each figure that were taken from Table V, whereas the solid blue lines are fits of the data by Eqs. (22) using T_A , labeled as T_{A^*} , as an adjustable parameter.

However, it is not a simple rigid-band shift. In addition to shifting to lower energy, these flat bands have much more broadening due to chemical disorder than other bands, which reflects the change in electron scattering length for states around E_F and alters transport properties, for example. An increase in the density of states (DOS) at E_F drives a magnetic instability according to the Stoner criterion^{75,76} that favors long-range order.

As our experimental data indicate, there is non-Fermi-liquid behavior in the alloying range of 20–30% Ni, which

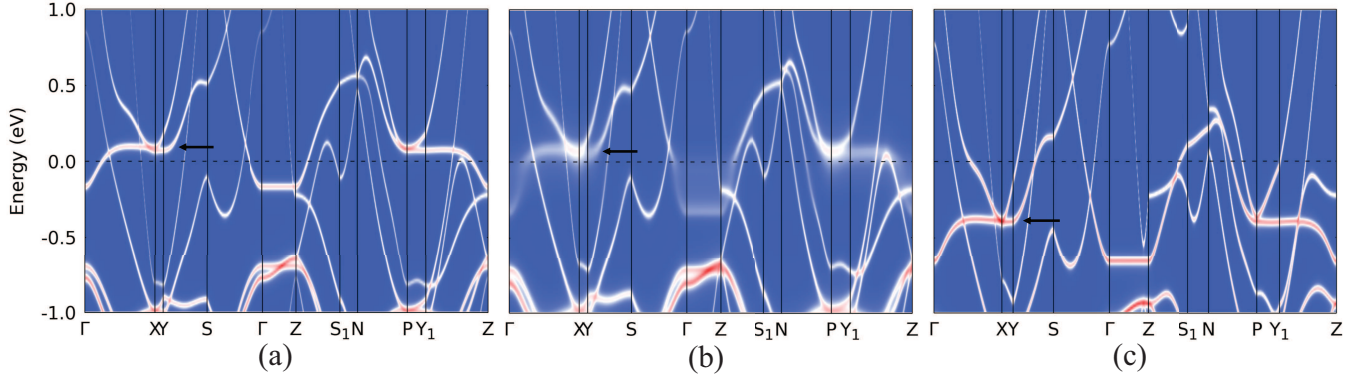


FIG. 18: KKR-CPA electronic dispersions (spectral functions) calculated for nonmagnetic $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$, at x values of (a) 0, (b) 0.15 and (c) 1. Color-coded intensity scale is high (red), medium (white), and low (blue), with flat bands (black arrows) indicated along $\Gamma - X - Y$ and $P - Y_1 - Z$ directions.

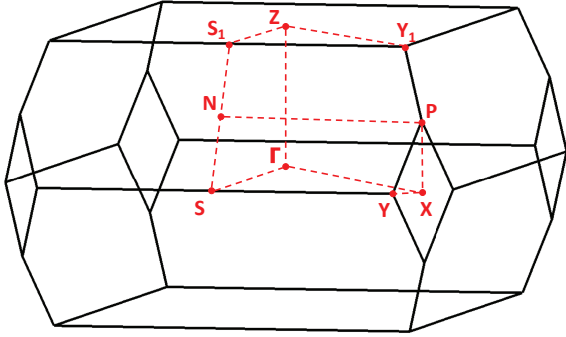


FIG. 19: Irreducible Brillouin zone of the body-centered tetragonal primitive unit cell of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ labeled with high-symmetry points and lines as used in Fig. 18.

coincides with the region of large dispersion broadening near E_F from chemical disorder found in KKR-CPA Bloch spectral functions, see Fig. (18), where, for ~ 20 – 30% Ni, k is a “less-good” quantum number for those Co $d_{x^2-y^2}$ states, with much shorter electron-scattering length. Then, above 30% Ni, the dispersion becomes dramatically less broadened as those states move below E_F , when k again becomes a “good” quantum number, as expected for a Fermi liquid. This effect is reflected in the DOS, see below, and therefore the heat capacity (next section).

For example, for pure SrNi_2As_2 [$x = 1$, Fig. 18(c)], the flat bands of Co $d_{x^2-y^2}$ character associated with magnetic instability are now far below E_F . Thus, in the Ni-rich region, the paramagnetic state (randomly-oriented finite local moments) prevails, while in the Co-rich region, the flat bands just above E_F can drive long-range magnetic order from electron doping upon substitution of a small amount of Ni for Co. Yet, the total energy difference between nonmagnetic (NM), ferromagnetic (FM) and A-type antiferromagnetic (AFMA) states is very small (~ 0.1 meV/f.u.) with a magnetic moment of $0.3 \mu_B$ at the Co sites in the magnetically-ordered

cases.

From the electronic dispersion in Fig. 18(a), the total density of states (DOS) of nonmagnetic SrCo_2As_2 calculated from KKR is plotted versus energy in Fig. 20(a). To validate our DOS results, the DOS calculated from a plane-wave basis set with the projector-augmented-wave (PAW) method in VASP^{77,78} on two \mathbf{k} -point meshes are plotted for comparison. The DOS curves over a large energy range agree including the peak just above the Fermi energy (E_F) due to the flat band of Co $d_{x^2-y^2}$ character. The $\text{DOS}(E_F)$ value of 11 states/(eV f.u.) including both spin directions calculated on the dense ($20 \times 20 \times 8$) \mathbf{k} -point mesh agrees with the previous study²⁸. With the additional enhancements from electron-phonon coupling and many-body renormalization of the band structure, this value can be compared directly with the result from specific heat measurements. In KKR-CPA, in order to control computational cost, we used the ($10 \times 10 \times 4$) \mathbf{k} -point mesh with a Gaussian smearing of 0.04 eV, which already gives good total energy convergence, but the $\text{DOS}(E_F)$ value becomes smaller from this numerical device. [Increasing the \mathbf{k} -mesh size improves the results for $\text{DOS}(E_F)$, as shown in Fig. 20(a).] However, the KKR-CPA result for $\text{DOS}(E_F)$ does reflect correct changes due to chemical disorder upon alloying with Ni and shows an interesting behavior. As seen in Fig. 20(b) which shows the DOS versus energy zoomed in around E_F for the compositions from $x = 0.0$ to 0.5, the main peak due to the flat band of Co $d_{x^2-y^2}$ character is increasingly broadened as x increases due to chemical disorder. At the same time, the DOS below E_F (at $E_F - 0.2$ eV) increases. For $\text{DOS}(E_F)$, the change is not monotonic; it first increases with alloying and reaches a maximum at $x = 0.2$, while the main DOS peak is still well above the Fermi energy, then it decreases upon further alloying. This behavior agrees with the trend in specific-heat measurements (see below) and shows that the chemical-disorder effects on the electronic dispersion do not correspond to a rigid-band-shifting picture.

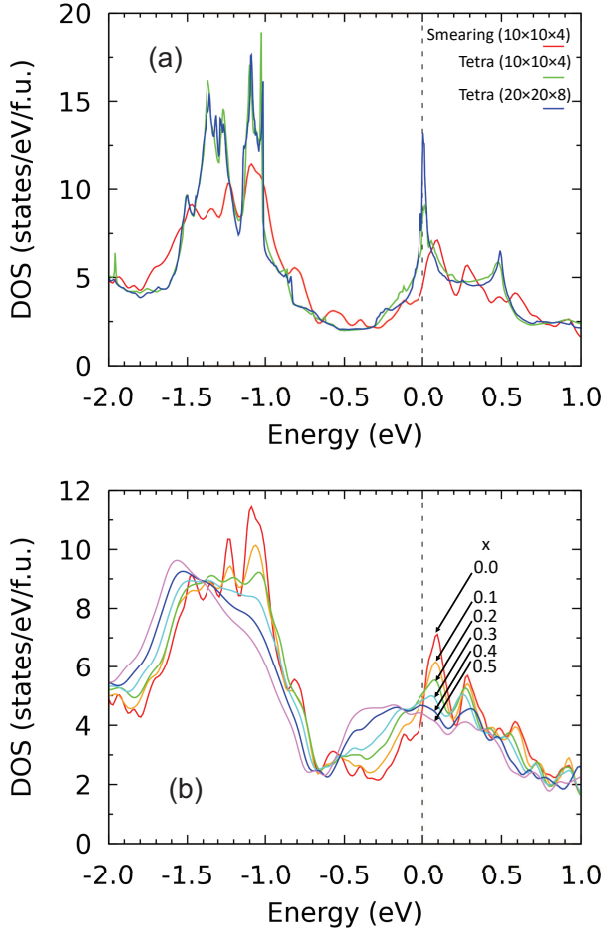


FIG. 20: (a) Electronic density of states (DOS) including both spin directions versus energy of nonmagnetic SrCo_2As_2 calculated in KKR with a Gaussian smearing of 0.04 eV on a \mathbf{k} -point mesh of $(10 \times 10 \times 4)$ in comparison to the DOS calculated with the tetrahedron method on \mathbf{k} -point meshes of $(10 \times 10 \times 4)$ and $(20 \times 20 \times 8)$, respectively. (b) KKR-CPA DOS including both spin directions versus energy calculated for nonmagnetic $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with compositions from $x = 0.0$ to 0.5. The Fermi energy E_F is at energy zero. The $\text{DOS}(E_F)$ first increases then decreases with increasing x .

VII. HEAT CAPACITY

A. Experimental Data and Fits

Figure 21 shows the heat capacity C_p versus T for the $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with compositions $x = 0, 0.04, 0.06, 0.10, 0.15, 0.20, 0.25, 0.30,$ and 0.50 over the entire T range of the measurements from 1.8 to 300 K in zero applied magnetic field. The $C_p(T)$ values at 300 K of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ are ≈ 125 J/mol K which is close to the classical Dulong-Petit value $C_V = 3nR = 124.7$ J/mol K, where $n = 5$ is the number of atoms per f.u. and R is the molar gas constant.

The low-temperature C_p/T vs T^2 data are shown in

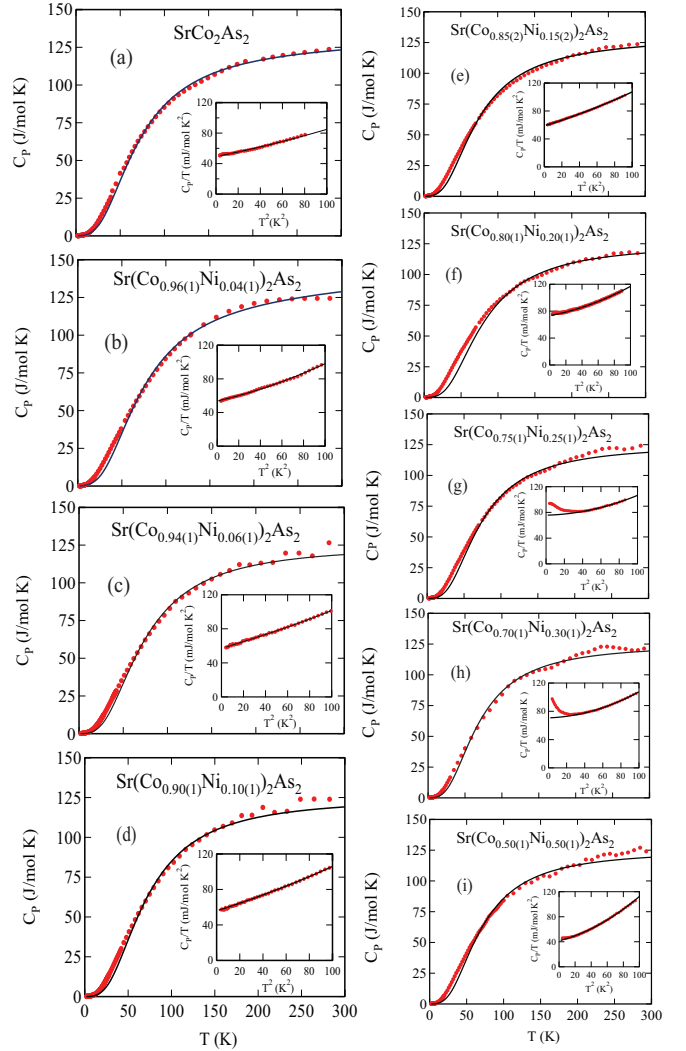


FIG. 21: Heat capacity C_p for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0, 0.04, 0.06, 0.10, 0.15, 0.20, 0.25, 0.30,$ and 0.50 single crystals versus temperature T . The solid curves in the main panels are fits of the data by Eqs. (26). Insets: C_p/T vs T^2 below 10 K. The solid lines are fits by Eq. (24) where the T ranges of the fits are 1.8 K to 10 K for all x values except for $x = 0.20, 0.25,$ and 0.30 for which the fit range was 6 K to 10 K, where extrapolations of the fits to 1.8 K are shown. One sees upturns in the low-temperature data for $x = 0.20, 0.25,$ and 0.30 associated with proximity to a magnetic quantum critical point at $x \approx 0.30$.

the insets of Fig. 21. The data were fitted over the temperature ranges given in the figure caption by

$$\frac{C_p(T)}{T} = \gamma + \beta T^2 + \delta T^4, \quad (24)$$

which is the sum of the electronic (γ) and low- T lattice ($\beta T^2 + \delta T^4$) heat capacity contributions. We fitted the data for $x = 0.20, 0.25$ and $x = 0.30$ only to illustrate the presence of low- T upturns in the data and these fits are not used henceforth. The fitted values of the Sommerfeld coefficient γ and the lattice-vibration parameters β and δ

for $x = 0$ to 0.15 and for $x = 0.50$ are listed in Table VII. The Debye temperature Θ_D is obtained from the value of β according to

$$\Theta_D = \left(\frac{12\pi^4 Rn}{5\beta} \right), \quad (25)$$

where n is the number of atoms per formula unit ($n = 5$ here). The values of Θ_D obtained from the β values are listed in Table VII.

The $C_p(T)$ data over the temperature range 100 to 280 K were fitted for $x = 0$ to 0.15 and for $x = 0.50$ by⁷⁹

$$C_p(T) = \gamma T + nC_{V\text{Debye}}(T), \quad (26)$$

$$C_{V\text{Debye}}(T) = 9R \left(\frac{T}{\Theta_D} \right)^3 \int_0^{\Theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} dx,$$

where $n = 5$ is the number of atoms per formula unit and $C_{V\text{Debye}}(T)$ is the Debye lattice heat capacity at constant volume per mole of atoms. The γ values in Table VII obtained from the low- T fits were used. The fits are shown as the black solid curves in Fig. 21 and the fitted values of the Debye temperature Θ_D are listed in Table VII. One sees that the values of Θ_D obtained from the fits by Eqs. (24) and (26) are reasonably close to each other for $x = 0$ to 0.15 and for $x = 0.50$.

The density of degenerate conduction carrier states at the Fermi energy E_F for both spin directions $D_\gamma(E_F)$ is obtained from γ according to

$$D_\gamma(E_F) = \frac{3\gamma}{\pi^2 k_B^2} \quad (27a)$$

which gives

$$D_\gamma(E_F) \left[\frac{\text{states}}{\text{eV f.u.}} \right] = \frac{1}{2.359} \gamma \left[\frac{\text{mJ}}{\text{mol K}^2} \right], \quad (27b)$$

where ‘‘mol’’ refers to a mole of formula units (f.u.). The $D_\gamma(E_F)$ values calculated for our $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals from their γ values using Eq. (27b) are listed in Table VII.

B. Non-Fermi-Liquid Types of Behavior in the Heat Capacity at Low Temperatures

As seen in the insets of Fig. 21, the $C_p(T)/T$ vs T^2 plots at low T show linear (Fermi-liquid) types of behavior for $x = 0.0, 0.04, 0.06, 0.10, 0.15$, and 0.50. However, for $x = 0.20, 0.25$, and 0.30, $C_p(T)/T$ vs T^2 departs from linearity below 10 K. In particular, the data for the latter two compositions exhibit clear upturns with decreasing T . The upturns in $C_p(T)/T$ at low T for $x = 0.25$ and 0.30 cannot result from high- T tails of Schottky anomalies arising from local-moment paramagnetic impurities because in that case the size of the upturns would *increase* with field, rather than decrease as demonstrated in Figs. 22(a)–(c). Similarly, in a metallic spin glass such

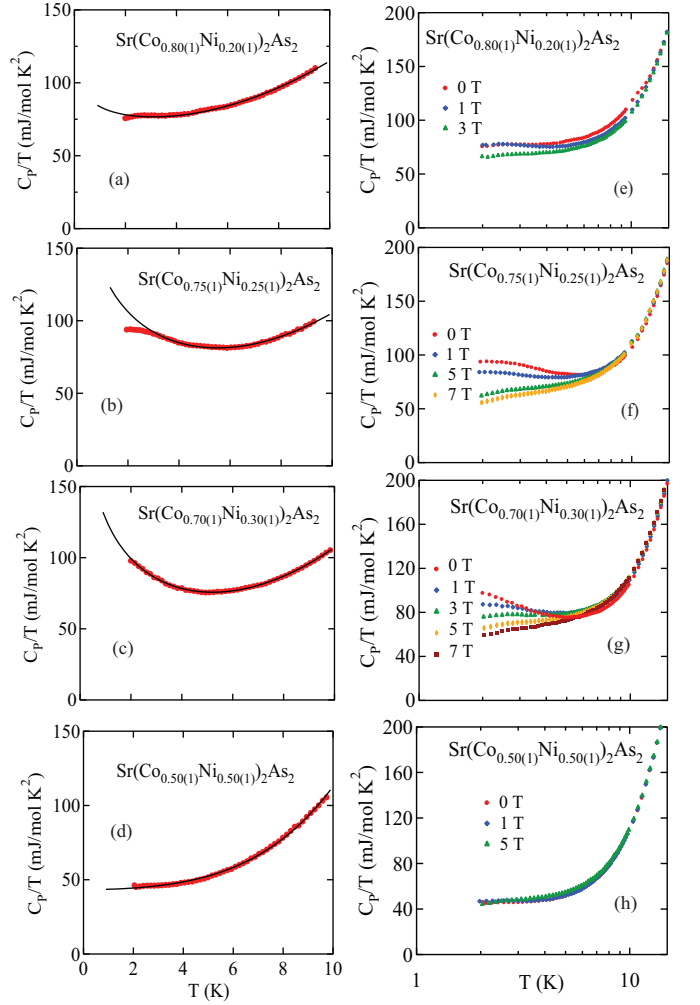


FIG. 22: (a)–(d) Heat capacity C_p/T vs T data at zero field from 1.8 to 10 K for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0.20, 0.25, 0.30$, and 0.50, respectively. The solid black curves in (a)–(c) are fits by Eq. (28). The data and fit in (d) are taken from the data and fit in the inset of Fig. 21(f). (e)–(h) C_p/T vs T at various fields on logarithmic temperature axes.

as Cu:Mn containing 2790 ppm of Mn and with a spin-glass transition temperature of 3.9 K, a 7 T field strongly *increases* the magnitude of the high- T tail of the broad peak in the heat capacity⁸⁰.

Because FM interactions dominate AFM interactions in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ for compositions of interest here as shown in previous sections, we fitted the $C_p(T)/T$ versus T data for $x = 0.20, 0.25$, and 0.30 from our minimum temperature of 2 K to 10 K by a model for FM quantum-critical behavior. For a 3D ferromagnet in the classical temperature regime at the QCP composition⁸¹, or for a 2D ferromagnet treated using scaling theory⁸², a contribution $\ln T$ to C/T is found, yielding the fit function

$$\frac{C_p(T)}{T} = \gamma + \beta T^2 + \delta T^4 + \kappa \ln(T/T_{\text{SF}}), \quad (28)$$

where γ and $\{\beta, \delta\}$ again represent the coefficients of

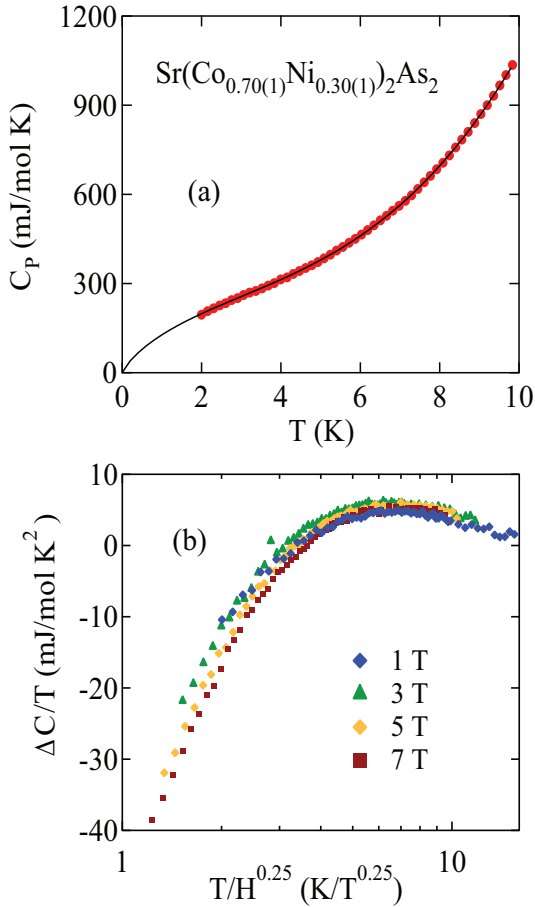


FIG. 23: (a) Heat capacity C_p vs temperature T in zero magnetic field from 1.8 to 10 K for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with the approximate quantum-critical composition $x = 0.30$. The solid black curve is a fit to the data using Eq. (28) and the parameters in Table VIII, with an extrapolation of the fit to zero temperature. (b) Scaling plot of the $\Delta C/T$ data in Fig. 22(g) versus $\log_{10}(T/H^{0.25})$.

the degenerate electronic and lattice heat-capacity contributions, respectively, κ represents the strength of the FM spin fluctuations, and T_{SF} is a characteristic spin-fluctuation temperature. The $C/T \sim \ln T$ contribution has been observed for quantum-critical fluctuations in the layered ferromagnet $\text{YFe}_2\text{Al}_{10}$ ⁸² and in the 3D ferromagnet $\text{Pd}_{1-x}\text{Ni}_x$ ⁸³. We find that Eq. (28) fits our low-temperature C_p/T versus T data for $x = 0.20, 0.25$, and 0.30 at low temperatures reasonably well, as shown in Figs. 22(a), 22(b), and 22(c), respectively, where the composition $x = 0.30$ is close to the quantum-critical value. The fitting parameters γ , β , δ , κ , T_{SF} , and the calculated values of Θ_{D} and $\mathcal{D}_\gamma(E_{\text{F}})$ obtained from β and γ utilizing Eqs. (25) and (27b), respectively, are listed in Table VIII.

If the quantum-critical behavior for $x = 0.30$ is indeed due to FM fluctuations, one would expect them to be suppressed by applied magnetic fields^{37,82}. Such was found to be the case, as shown in Fig. 22(g). After subtracting

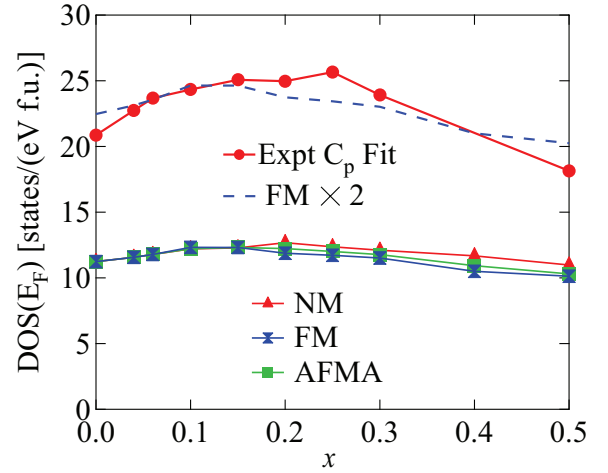


FIG. 24: KKR-CPA electronic density of states at the Fermi energy [$\text{DOS}(E_{\text{F}})$] for both spin directions calculated for nonmagnetic (NM), ferromagnetic (FM) and A-type antiferromagnetic (AFMA) $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with compositions $x = 0.0$ to 0.5 . The experimental densities of states $\text{DOS}(E_{\text{F}})$ (Expt C_p Fit) in Tables VII and VIII obtained from the electronic heat capacity coefficients γ using Eq. (27b) are plotted for comparison. The blue dashed line is the theoretical FM $\text{DOS}(E_{\text{F}})$ versus x in the lower part of the figure multiplied by two.

the $H = 0$ data from the $H > 0$ data in Fig. 22(g), one obtains the quantity

$$\frac{\Delta C}{T} \equiv \frac{C_p(H)}{T} - \frac{C_p(H=0)}{T}, \quad (29)$$

which is plotted versus $\log_{10}(T/H^{0.25})$ in Fig. 23(b), where the H exponent was adjusted to obtain the best scaling. A reasonably good scaling is seen. A very good scaling of $\Delta C/T$ versus $\log_{10}(T/B^{0.59})$ was previously found for the quasi-2D ferromagnet $\text{YFe}_2\text{Al}_{10}$ in Ref.⁸², where the compound is found to be intrinsically quantum-critical.

C. Comparison of Calculated Electronic-Structure DOS with Experimental Heat Capacity DOS

Figure 24 shows the KKR-CPA density of states (DOS) at E_{F} versus electron doping x for the nonmagnetic (NM), ferromagnetic (FM), and A-type antiferromagnetic (AFMA) states, with similar results. The DOS is seen to first increase up to $x \approx 0.2$ and then slowly decrease as a function of Ni substitution x on the Co site (electron-doping), while the main DOS peak is still above E_{F} [green line in Fig. 20(b)]. This change in the $\text{DOS}(E_{\text{F}})$ results from the significant broadening of the flat bands around E_{F} . The variation in $\text{DOS}(E_{\text{F}})$ with x is compared in Fig. 24 with the results obtained from the experimental $\text{DOS}(E_{\text{F}})$ values in Tables VII and VIII obtained by fitting the heat capacity data. One sees

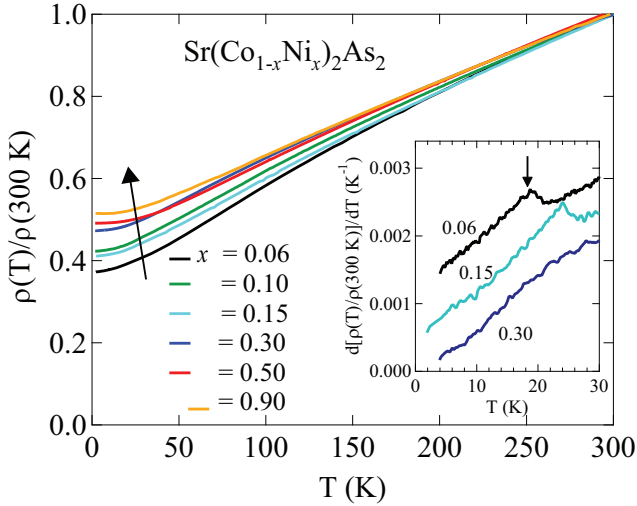


FIG. 25: Normalized electrical resistivity $\rho(T)/\rho(300\text{ K})$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with $x = 0.06, 0.10, 0.15, 0.30, 0.50,$ and 0.90 . The inset shows the temperature derivative of the normalized resistivity in the vicinity of T_N . The derivative curves for $x = 0.06$ and 0.15 reveal sharp features at magnetic transitions (indicated with an arrow for $x = 0.06$) and smooth evolution without sharp features in $x = 0.30$.

that the x dependence of the observed and calculated $\text{DOS}(E_F)$ types of behavior are very similar, but the observed values obtained from the heat capacity are shifted upwards. This difference can be accounted for by the presence of an electron-phonon coupling constant λ_{ep} and a many-body renormalization of the band structure via electron-electron interactions represented by an electron-electron coupling constant λ_{ee} according to²⁶

$$\text{DOS}(E_F)^{\text{obs}} = \text{DOS}(E_F)^{\text{calc}}(1 + \lambda_{ep})(1 + \lambda_{ee}), \quad (30)$$

where typically $0 < \lambda_{ep} \lesssim 1$. We infer from Fig. 24 that the net enhancement of the band-structure density of states is about a factor of two, as shown by the dashed blue line in Fig. 24. This enhancement is about the same as in BaCo_2As_2 ²⁸. Hence, the behavior is explained well by the disorder-induced effects that lead to competing magnetic states (NM, FM, and AFMA), see Sec. VID.

VIII. ELECTRICAL RESISTIVITY

The in-plane electrical resistivity of the $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals was studied for compositions with $x = 0.06, 0.10, 0.15, 0.30, 0.50,$ and 0.90 . All compositions show metallic temperature-dependent resistivity in zero applied field, as shown in Fig. 25. The resistivity value at room temperature was determined to be $\approx 120 \pm 30 \mu\Omega\text{ cm}$ as seen in Fig. 26(a), and did not show any systematic trend with x . The large error bars reflect the fact that the measurements were not performed on a statistically significant array of samples

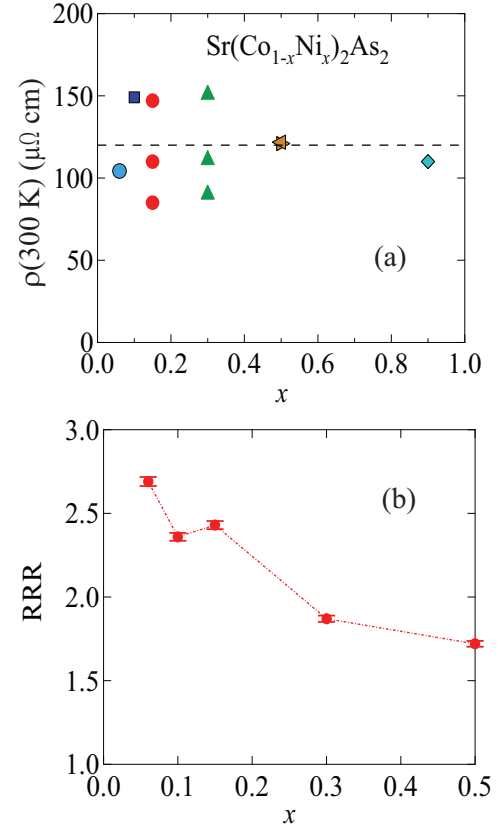


FIG. 26: (a) Electrical resistivity ρ at $T = 300\text{ K}$ and (b) residual resistivity ratio $\text{RRR} \equiv \rho(2\text{ K})/\rho(300\text{ K})$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with $x = 0.06, 0.10, 0.15, 0.30,$ and 0.50 .

of each composition and are known to suffer from potential cracks⁸⁴ affecting the correct determination of the sample geometric factor. Nonetheless, this behavior is notably different from the evolution of the resistivity at room temperature in electron-doped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, which shows a factor of three decrease in a similar doping range⁸⁵. The behavior in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ is rather similar to the doping-independent value of $\rho(300\text{ K})$ found in hole-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ⁸⁶. To exclude the effect of a systematic error in the comparison of quantitative resistivity measurements, we analyze all resistivity data using normalized $\rho(T)/\rho(300\text{ K})$ values.

The expected anomalies in the temperature-dependent resistivity at the magnetic transitions⁸⁷ are very weak in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ and can be noticed only in the derivative plots, $d\rho(T)/dT$, shown in the inset of Fig. 25. Note that the derivative exhibits sharp peaks at around 20 K at the respective Néel temperatures for $x = 0.06$ and $x = 0.15$. The derivative for the $x = 0.30$ crystal with no magnetic ordering shows no such maximum. The resistivity in Fig. 25 at low temperatures is determined mostly by substitutional disorder, with the residual resistivity ratio $\rho(300\text{ K})/\rho(1.8\text{ K})$ in Fig. 26(b) decreasing from 20 in pure SrCo_2As_2 ²⁸ to 2.7 for $x = 0.06$ and 1.8

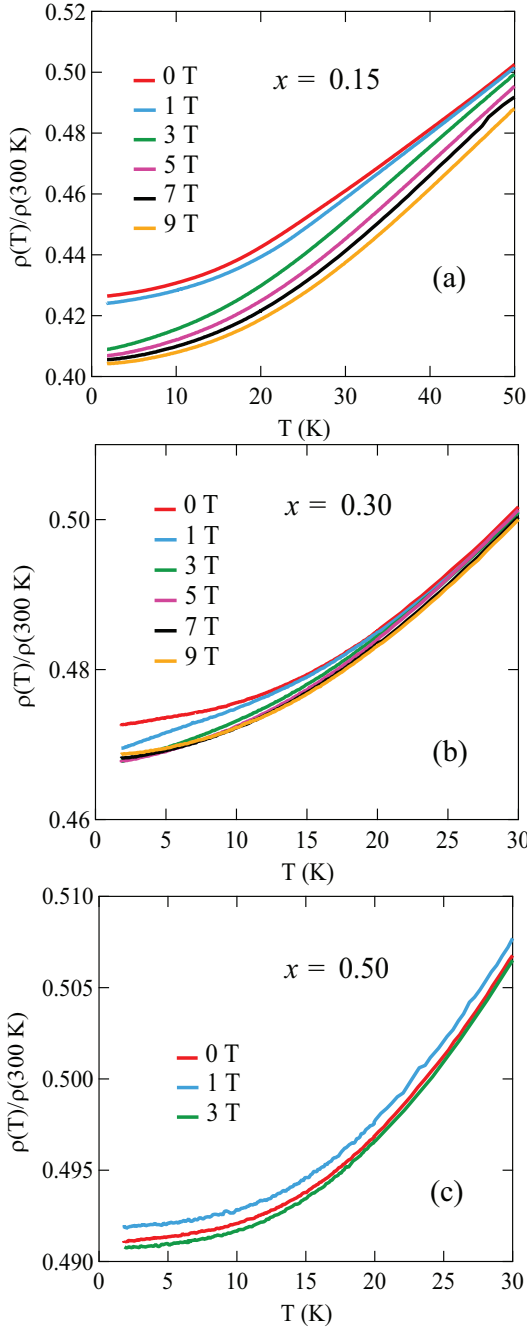


FIG. 27: In-plane normalized resistivity $\rho(T)/\rho(300\text{ K})$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with compositions $x = 0.15, 0.30,$ and 0.50 measured in the indicated magnetic fields $H \parallel c$ as a function of temperature T .

for $x = 0.50$.

The resistivity notably decreases on application of a magnetic field parallel to the c -axis as shown in Fig. 27. This negative magnetoresistance clearly shows a contribution of spin-disorder scattering in the samples. A more detailed inspection of the low-temperature part of the temperature-dependent resistivity is made using ρ versus T^2 plots as shown in Fig. 28. In such plots the

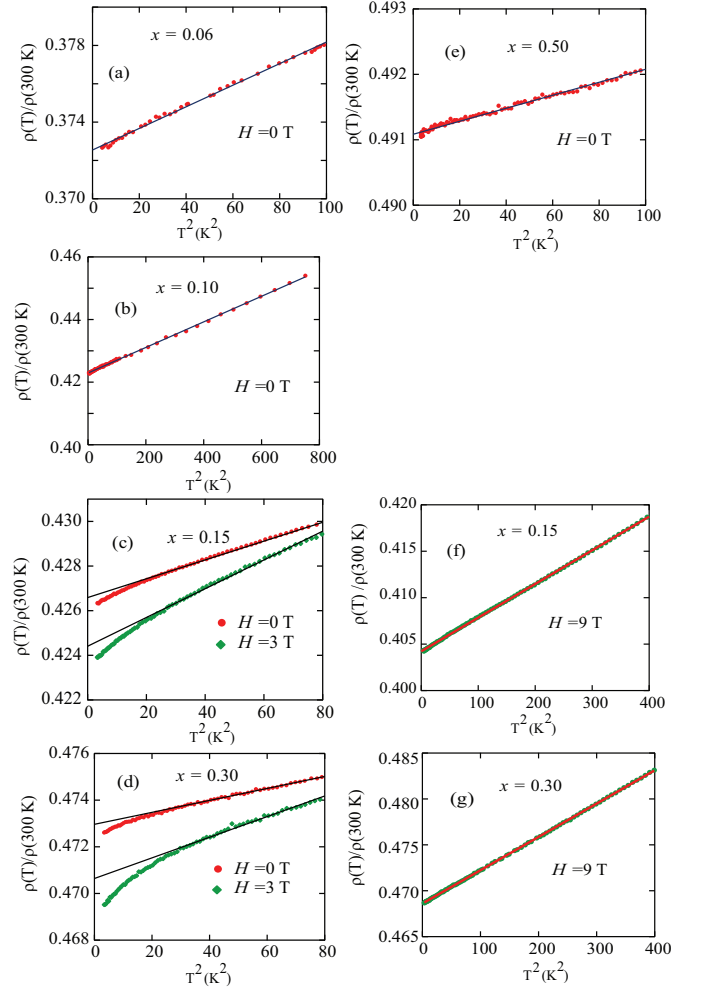


FIG. 28: (a)–(e) In-plane normalized resistivity $\rho(T)/\rho(300\text{ K})$ vs T^2 for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with compositions $x = 0.06, 0.10, 0.15, 0.30,$ and 0.50 in magnetic fields $H_c = 0$ and/or 3 T , as indicated. (f, g) $\rho(T)/\rho(300\text{ K})$ vs T^2 for $x = 0.15$ and 0.30 in $H_c = 9\text{ T}$.

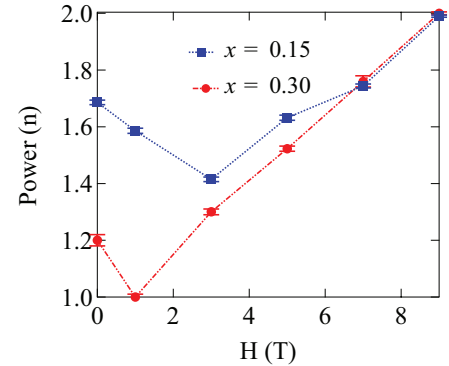


FIG. 29: The power n in the expression $\rho(T) = \rho_0 + AT^n$ for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals with $x = 0.15$ and 0.30 at low temperatures $1.8\text{ K} \lesssim T \lesssim 9\text{ K}$ versus applied magnetic field H_c . These fits are not related to the linear fits shown in Fig. 28.

temperature-dependent resistivity expected for a Fermi liquid, $\rho = \rho(0) + AT^n$ with $n = 2$, would be a straight line. This is indeed observed in zero magnetic field for the crystals with $x = 0.06, 0.10$, and 0.50 in panels (a), (b), and (e), respectively, consistent with the heat capacity data in the respective insets of Fig 21.

For crystals with $x = 0.15$ and 0.30 , downward deviations from the linear dependence are found in Fig. 28 at the lowest temperatures, suggesting that the exponent n of the power-law function becomes smaller than 2, thus revealing non-Fermi-liquid behavior^{88,89}. The exponent n found from fits to the resistivity data between 1.8 K and 9 K (not shown) exhibits a notable dependence on magnetic field (see Fig. 29), as is observed in systems with magnetic-field-tuned quantum-critical points such as YbRh₂Si₂⁹⁰, CeCoIn₅⁹¹, YbAgGe⁹², and YbPtBi⁹³. The minimum in the $n(H)$ dependence is observed at 3 T for the crystal with $x = 0.15$ and at 1 T for $x = 0.30$. In the latter case the exponent at the $n(H)$ minimum is close to $n = 1$, as observed in cuprates, organics and heavy fermion systems at their quantum critical points⁹⁴. The fact that the characteristic field at which the exponent n is minimum decreases with increasing x suggests that quantum-critical behavior may be observed in zero field for a composition near $x = 0.30$, as we observe in the heat capacity.

IX. DISCUSSION OF NON-FERMI-LIQUID TYPES OF BEHAVIOR IN Sr(Co_{1-x}Ni_x)₂As₂

Non-Fermi-liquid (NFL) types of behavior in d - and f -band metals have been intensively studied since the 1990's^{88,89}, where the early studies emphasized theoretical and experimental studies of compounds containing lanthanides and/or uranium. Studies of d -band compounds other than those presented here have previously been reported, such as studies of ruthenates⁹⁵, high- T_c cuprates, and iron pnictides⁹⁶, where materials showing NFL characteristics are sometimes dubbed “bad metals” or “strange metals”. NFL behavior has been documented in P-doped 122-type iron arsenide crystals BaFe₂(As_{1-x}P_x)₂⁹⁷. For optimally hole-doped (Ba_{0.6}K_{0.4})Fe₂As₂ crystals with a superconducting onset temperature $T_c = 39$ K, high-resolution laser-based angle-resolved photoemission spectroscopy (ARPES) measurements indicated that the normal state of this material has a well-defined Fermi surface but no quasiparticles⁹⁸. Furthermore, the superconducting condensate is found to be coherent but the temperature dependence of the superconducting gap dramatically differs from the Bardeen-Cooper-Schrieffer (BCS) prediction⁹⁸. Such types of behavior originate from criticality⁹⁹, where quantum-mechanical fluctuations drive phase transitions at $T = 0$ upon varying a control parameter such as pressure, magnetic field, or a material's composition, but quantum criticality can also have important observable finite-temperature manifestations^{89,99}.

A special case of this phenomenon is the *local* marginal Fermi liquid as originally applied to the high- T_c cuprates, in which the energy scale for the low-energy particle-hole excitations is set by the temperature^{100,101}. The adjectives “local” or “nonlocal” in the context of marginal Fermi liquids refer to whether the effective interactions between quasiparticles are of short or long range, respectively¹⁰². Some of the predicted properties are in agreement with measurements on these cuprates, such as the T -linear normal-state electrical resistivity over a wide range of temperature above T_c , in contrast to the T^2 behavior expected for a Fermi liquid, and the optical conductivity. For our Sr(Co_{1-x}Ni_x)₂As₂ crystals, an example of $\rho \sim T^n$ behavior with $n = 1$ was found for $x = 0.30$ in $H = 1$ T (see Fig. 29); for other fields with $0 \leq H \leq 7$ T we obtained $1.2 \leq n \leq 1.8$ for both $x = 0.15$ and 0.30 , but with both crystals showing Fermi-liquid behavior with $n = 2$ at the higher field $H = 9$ T.

Itinerant weak ferromagnetism was discovered in a polycrystalline sample of ZrZn₂ in 1957¹⁰³. This was surprising because both Zr and Zn are superconducting and are nonmagnetic elements. The Curie temperature and the low- T saturation moment of their polycrystalline sample were estimated as $T_C = 35$ K and $\mu_{\text{sat}} = 0.13 \mu_B/\text{f.u.}$, respectively¹⁰³. Subsequent work on high-quality single crystals of ZrZn₂^{102,104-107} yielded $T_C = 28.5$ K and $\mu_{\text{sat}} = 0.17 \mu_B/\text{f.u.}$ ¹⁰⁵. Here $\rho \propto T^{5/3}$, attributed to the presence of a *nonlocal* marginal Fermi liquid state in ZrZn₂¹⁰². Our $\rho(T)$ data in Fig. 29 for a Sr(Co_{1-x}Ni_x)₂As₂ crystal with $x = 0.15$ has a temperature exponent $n \sim 5/3$ for $H = 0, 1, 5$, and 7 T, and for $x = 0.30$ at $H = 7$ T. We note that the Sommerfeld coefficient of ZrZn₂ in $H = 0$ is large, $\gamma = 45$ mJ/mol K²¹⁰⁴, of the same order as the values for Sr(Co_{1-x}Ni_x)₂As₂ in Tables VII and VIII. However, the $C_p(T)$ data for ZrZn₂¹⁰⁶ do not show the clear upturns we see for Sr(Co_{1-x}Ni_x)₂As₂ crystals with $x = 0.25$ and 0.30 in the insets of Fig. 21 and in Fig. 22 because ZrZn₂ is not close to a quantum critical point. On the other hand, $C_p(T)$ at low T for ZrZn₂ is suppressed with increasing H ¹⁰⁴ as we also observe in Fig. 22, which likely arises from the suppression of FM fluctuations with field in both systems. The low- T resistivity of ZrZn₂ increases with increasing H ¹⁰⁶, whereas we observe a decrease in the normalized resistivity for $x = 0.15$ and 0.30 in Figs. 27 and 28.

Thus there appears to be a field- and composition-dependent crossover in the nature of the non-Fermi liquid when it occurs in the Sr(Co_{1-x}Ni_x)₂As₂ system.

X. SUMMARY

The physical properties of Ni-doped Sr(Co_{1-x}Ni_x)₂As₂ single crystals were investigated for compositions $x = 0$ to 0.9 . Single-crystal x-ray diffraction studies at room temperature showed a continuous crossover from the ucT (uncollapsed tetragonal) to cT (collapsed tetrago-

nal) structure with increasing x . The temperature T dependence of the magnetic susceptibilities χ_{ab} and χ_c demonstrated that the paramagnetic (PM) ground state of SrCo_2As_2 transforms to an antiferromagnetic (AFM) state with only a trace amount of Ni doping ($x = 0.013$). This appears to be consistent with our *ab initio* electronic-structure calculations for $x = 0$ that exhibit a flat-band peak in the density of states just above the Fermi energy that contributes to magnetic ordering at small doping levels. The AFM order persists up to $x = 0.25$, above which the PM state is observed for $x = 0.30$ and $x = 0.50$.

Based on molecular field theory (MFT) for local magnetic moments, the $\chi(T)$ data at temperatures T less than the Néel temperature T_N indicate that the ordered moments are aligned within the ab plane. The nonzero limits of $\chi_{ab}(T < T_N)$ suggest a planar helical AFM structure. We calculated the turn angle kd between adjacent ab -plane ordered moments along the helix c -axis within MFT from the ratio $\chi_{ab}(T = 0)/\chi(T_N)$ for the crystals showing AFM ordering and found that the helix turn angle varies with Ni concentration. Magnetic neutron diffraction measurements confirm the helical magnetic structure with turn angles of the same order as those estimated here.

The $\chi^{-1}(T)$ data at $T > T_N$ follow Curie-Weiss-like behavior. The Weiss temperature and T_N follow the same trend, initially increasing and then decreasing with increasing Ni doping. The positive Weiss temperatures in the range $x = 0.04 - 0.25$ indicate that ferromagnetic exchange interactions are dominant in these AFM crystals. In terms of our helical AFM structure hypothesis, the positive Weiss temperatures reflect the FM intralayer interactions needed to line up the spins within a layer of the helix, and that these are significantly stronger than the interlayer interactions which must include AFM interactions to drive the AFM order.

The magnetization versus field $M(H)$ data below T_N with \mathbf{H} in the ab plane exhibit a spin-flop-like transition followed by a second-order transition to the PM state for $x = 0.04$ to 0.20 , whereas c -axis measurements reveal only the expected second-order AFM to PM transition. The saturation moment in the ordered state increases initially with Ni doping and reaches a maximum at $0.10 < x < 0.15$, then decreases for $x = 0.25$ followed by paramagnetic $M(H)$ behavior for $x \geq 0.30$.

The ordered moment μ_{sat} per transition-metal atom in the AFM state obtained by extrapolating the high-field $M(H)$ data to zero field is small relative to the effective moment μ_{eff} in the PM state, indicating that $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ is essentially an itinerant ferromagnetic system. In addition, the magnetic moment per formula unit versus applied field H does not saturate at high fields up to 14 T. Instead, at the higher fields it increases approximately linearly with field as often observed in weak itinerant FMs. As noted above, the crystals are AFM in zero field with the ordered moments oriented within the ab plane. However, from the Weiss

temperature in the Curie-Weiss law at $T > T_N$ the dominant interactions are FM.

The maximum values of the ab -plane spin-flop-like field and the ab -plane critical field $H_{c,ab}$ at which the PM state is entered are both less than about 4 T for all $T < T_N(x)$. Therefore our isothermal $M_{ab}(H)$ data measured up to 14 T were analyzed in terms of Takahashi's spin-fluctuation theory for weak itinerant ferromagnets via M^2 vs H/M and M^4 vs H/M isotherms at different temperatures for fields above about 4 T. The linear behavior of the M^4 versus H/M isotherm at high fields for $T \approx T_N$ (i.e., at the Curie temperature T_C within Takahashi's theory) observed for $x = 0.10, 0.15$, and 0.20 indicates that the critical magnetic isotherm at T_C is strongly influenced by thermally-induced ferromagnetic spin fluctuations.

Takahashi's spin-fluctuation spectral parameter T_0 obtained from fitting the high-field $M_{ab}(H)$ measurements at $T = 2$ K and $T = T_C$ and the other spectral parameter T_A used as a fitting parameter yield an excellent fit of $\chi^{-1}(T > T_C)$ by Takahashi's theory for the normal-state inverse susceptibility, confirming that $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ acts like a weak itinerant ferromagnet for high fields $H_{ab} > 4$ T at $T < T_C$ and for small fields at $T > T_C$.

The heat capacity $C_p(T)$ data exhibit a Fermi-liquid behavior at low temperature for $x = 0, 0.04, 0.06, 0.10, 0.15$, and 0.50 . No clear feature in $C_p(T)$ was observed near T_N for any of the crystals that exhibit AFM ordering, as similarly found²⁶ for the itinerant AFM $\text{CaCo}_{2-y}\text{As}_2$. This absence evidently arises from the small entropy change at T_N associated with the itinerant nature of the magnetism. A low-temperature upturn in $C_p(H = 0, T)/T$ was observed for $x = 0.25$. This upturn was stronger for 0.30 which is near the quantum-critical concentration separating the AFM and PM phases. The upturns were fitted by a $\ln T$ temperature dependence consistent with ferromagnetic quantum-critical fluctuations. These low- T $C_p(T)/T$ upturns and hence the FM fluctuations were suppressed in a field $H = 7$ T as expected for ferromagnetic fluctuations. A scaling of the field-dependent heat capacity $\Delta C/T$ against $T/H^{0.25}$ was demonstrated, also suggestive of quantum-critical behavior.

Negative magnetoresistance was observed indicating a significant contribution of spin-disorder scattering. In most compositions the electrical resistivity at low temperatures follows a $\rho = \rho_0 + AT^n$ dependence with $n = 2$, as expected for a Fermi liquid. However, deviations with $n < 2$ are found for compositions $x = 0.15$ and 0.30 , the latter being near the boundary of the long-range magnetic-ordering composition that also show a notable dependence on magnetic field. These observations are consistent with the non-Fermi-liquid types of behavior observed in the heat capacity measurements and suggest the occurrence of a quantum critical point near $x = 0.30$.

In conclusion, the Ni doping in SrCo_2As_2 substantially affects the magnetic ground state and spin-spin correlations in $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$, causing a paramagnetic to

antiferromagnetic phase transition as well as the spin-fluctuation-mediated non-Fermi-liquid state near $x = 0.3$ where the long-range magnetic order disappears. The long-range order thus terminates at an unusual quantum critical point at $x \approx 0.3$ at which the ferromagnetic-interaction-dominated antiferromagnetic phase undergoes a transition to the paramagnetic phase.

APPENDIX: TABLES OF FITTED PARAMETERS

TABLE I: Crystallographic data for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ ($x = 0 - 1$) single crystals at room temperature, including the fractional c -axis position parameter z_{As} of the As atoms, the tetragonal lattice parameters a and c , the unit cell volume V_{cell} containing two formula units of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$, and the c/a ratio. The compositions in the first column were obtained from EDS analyses.

Compound	z_{As}	a (Å)	c (Å)	V_{cell} (Å ³)	c/a
SrCo_2As_2	0.35770(11)	3.955(3)	11.684(9)	182.76(9)	2.954(4)
$\text{Sr}(\text{Co}_{0.987(1)}\text{Ni}_{0.013(1)})_2\text{As}_2$	0.3574(2)	3.9601(16)	11.653(5)	182.74(17)	2.942(2)
$\text{Sr}(\text{Co}_{0.96(1)}\text{Ni}_{0.04(1)})_2\text{As}_2$	0.3579(3)	3.958(3)	11.667(12)	182.8(3)	2.948(5)
$\text{Sr}(\text{Co}_{0.94(1)}\text{Ni}_{0.06(1)})_2\text{As}_2$	0.35740(14)	3.957(6)	11.618(16)	181.9(6)	2.936(8)
$\text{Sr}(\text{Co}_{0.90(1)}\text{Ni}_{0.10(1)})_2\text{As}_2$	0.35759(14)	3.970(2)	11.566(7)	182.3(2)	2.913(3)
$\text{Sr}(\text{Co}_{0.85(2)}\text{Ni}_{0.15(2)})_2\text{As}_2$	0.35769(11)	3.976(5)	11.530(16)	182.3(5)	2.899(7)
$\text{Sr}(\text{Co}_{0.80(1)}\text{Ni}_{0.20(1)})_2\text{As}_2$	0.35799(13)	4.005(6)	11.482(16)	184.2(6)	2.867(8)
$\text{Sr}(\text{Co}_{0.75(1)}\text{Ni}_{0.25(1)})_2\text{As}_2$	0.3581(4)	4.002(2)	11.437(6)	183.2(2)	2.858(2)
$\text{Sr}(\text{Co}_{0.70(1)}\text{Ni}_{0.30(1)})_2\text{As}_2$	0.35799(10)	4.005(5)	11.420(15)	183.2(5)	2.851(7)
$\text{Sr}(\text{Co}_{0.60(1)}\text{Ni}_{0.40(1)})_2\text{As}_2$	0.35770(5)	4.028(2)	11.181(6)	181.4(2)	2.776(2)
$\text{Sr}(\text{Co}_{0.50(1)}\text{Ni}_{0.50(1)})_2\text{As}_2$	0.35775(7)	4.042(6)	11.030(18)	180.2(6)	2.729(8)
$\text{Sr}(\text{Co}_{0.40(1)}\text{Ni}_{0.60(1)})_2\text{As}_2$	0.35840(9)	4.074(5)	10.850(14)	180.1(5)	2.663(6)
$\text{Sr}(\text{Co}_{0.30(1)}\text{Ni}_{0.70(1)})_2\text{As}_2$	0.35886(5)	4.0921(15)	10.723(4)	179.55(15)	2.620(2)
$\text{Sr}(\text{Co}_{0.20(1)}\text{Ni}_{0.80(1)})_2\text{As}_2$	0.36010(4)	4.1154(8)	10.522(2)	178.21(8)	2.557(1)
$\text{Sr}(\text{Co}_{0.10(1)}\text{Ni}_{0.90(1)})_2\text{As}_2$	0.36147(4)	4.1366(9)	10.355(2)	177.19(9)	2.503(1)
$\text{SrNi}_2\text{As}_2^a$	0.36244(8)	4.1483(11)	10.231(3)	176.06(11)	2.466(1)

^agrown in Bi flux

TABLE II: Parameters obtained from Modified Curie-Weiss fits to $\chi(T)$ data between 100 and 300 K for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ crystals using Eq. (1). Shown are the T -independent contribution to the susceptibility χ_0 , Curie constant per mole of formula units C_α with the applied field in the $\alpha = ab, c$ directions, the Weiss temperature $\theta_{p\alpha}$, and the difference $\Delta\theta_p$ between the ab plane and c -axis Weiss temperatures $\Delta\theta_p \equiv \theta_{pab} - \theta_{pc}$. The effective Bohr magneton number per transition metal atom (Co and/or Ni) $p_{\text{eff}\alpha}$ was calculated from C_α using Eq. (4).

Compound	Field Orientation	χ_0 ($10^{-4} \frac{\text{cm}^3}{\text{mol f.u.}}$)	C_α ($\frac{\text{cm}^3\text{K}}{\text{mol f.u.}}$)	$p_{\text{eff}\alpha}$	$\theta_{p\alpha}$ (K)	$\Delta\theta_p$ (K)
SrCo_2As_2	$H \parallel ab$	-4.2(2)	0.92(1)	1.92(1)	-128(3)	-39(4)
	$H \parallel c$	-6.1(2)	1.12(1)	2.12(1)	-167(3)	
$\text{Sr}(\text{Co}_{0.987(1)}\text{Ni}_{0.013(1)})_2\text{As}_2$	$H \parallel ab$	1.7(1)	0.442(4)	1.329(6)	-6.4(9)	-0.5(9)
	$H \parallel c$	1.9(2)	0.438(5)	1.323(7)	-6.9(1)	
$\text{Sr}(\text{Co}_{0.96(1)}\text{Ni}_{0.04(1)})_2\text{As}_2$	$H \parallel ab$	6.6(8)	0.441(3)	1.328(4)	10.4(6)	4.5(7)
	$H \parallel c$	6.3(8)	0.466(2)	1.365(3)	5.9(4)	
$\text{Sr}(\text{Co}_{0.94(1)}\text{Ni}_{0.06(1)})_2\text{As}_2$	$H \parallel ab$	5.4(2)	0.385(1)	1.241(1)	23.1(2)	6.0(5)
	$H \parallel c$	5.4(1)	0.419(3)	1.294(5)	17.1(4)	
$\text{Sr}(\text{Co}_{0.90(1)}\text{Ni}_{0.10(1)})_2\text{As}_2$	$H \parallel ab$	6.8(3)	0.385(1)	1.241(1)	32.9(2)	1.7(4)
	$H \parallel c$	5.6(1)	0.411(2)	1.282(3)	31.2(3)	
$\text{Sr}(\text{Co}_{0.85(2)}\text{Ni}_{0.15(2)})_2\text{As}_2$	$H \parallel ab$	5.5(4)	0.359(1)	1.198(2)	35.2(3)	4.5(4)
	$H \parallel c$	5.68(3)	0.393(1)	1.253(1)	30.7(2)	
$\text{Sr}(\text{Co}_{0.80(1)}\text{Ni}_{0.20(1)})_2\text{As}_2$	$H \parallel ab$	5.55(6)	0.313(2)	1.119(3)	25.6(5)	4.2(3)
	$H \parallel c$	4.95(2)	0.360(1)	1.200(1)	21.4(1)	
$\text{Sr}(\text{Co}_{0.75(1)}\text{Ni}_{0.25(1)})_2\text{As}_2$	$H \parallel ab$	14.4(3)	0.245(1)	0.989(2)	17.6(3)	3.8(3)
	$H \parallel c$	12.7(3)	0.288(1)	1.073(2)	13.8(3)	
$\text{Sr}(\text{Co}_{0.70(1)}\text{Ni}_{0.30(1)})_2\text{As}_2$	$H \parallel ab$	5.14(3)	0.199(1)	0.892(2)	0.42(3)	1.7(3)
	$H \parallel c$	1.18(2)	0.219(1)	0.935(2)	-1.3(3)	
$\text{Sr}(\text{Co}_{0.50(1)}\text{Ni}_{0.50(1)})_2\text{As}_2$	$H \parallel ab$	7.8(2)	0.164(4)	0.81(1)	-83(3)	-3(4)
	$H \parallel c$	7.7(5)	0.187(5)	0.86(1)	-86(3)	

TABLE III: Parameters obtained from modified Curie-Weiss fits to $\chi(T)$ data between 100 and 300 K for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ using Eq. (1). Shown are the AFM transition temperature T_N , the angle-averaged Curie constant per mol $C_{\text{ave}} = (2C_{ab} + C_c)/3$, the average Weiss temperature $\theta_{p,\text{ave}} = (2\theta_{pab} + \theta_{pc})/3$, the angle-averaged effective Bohr magneton number per transition metal atom $p_{\text{eff,ave}}$ calculated from Eq. (4), and the ratio $f = \theta_{p,\text{ave}}/T_N$. Also listed is the turn angle kd between adjacent FM-aligned magnetic layers of a helix model obtained using Eq. (11).

Compound	T_N (K)	C_{ave} $\left(\frac{\text{cm}^3\text{K}}{\text{mol}}\right)$	$p_{\text{eff,ave}}$	$\theta_{p,\text{ave}}$ (K)	$f = \theta_{p,\text{ave}}/T_N$	kd (π rad)	kd (degree)
SrCo_2As_2		0.99(1)	1.99(1)	-141(2)			
$\text{Sr}(\text{Co}_{0.987(1)}\text{Ni}_{0.013(1)})_2\text{As}_2$	3.01(2)	0.441(4)	1.33(1)	-6.6(2)	-2.2		
$\text{Sr}(\text{Co}_{0.96(1)}\text{Ni}_{0.04(1)})_2\text{As}_2$	11.16(3)	0.449(3)	1.340(4)	8.9(5)	0.8	0.58	105
$\text{Sr}(\text{Co}_{0.94(1)}\text{Ni}_{0.06(1)})_2\text{As}_2$	20.8(6)	0.396(1)	1.259(2)	21.1(3)	1.01	0.52	93
$\text{Sr}(\text{Co}_{0.90(1)}\text{Ni}_{0.10(1)})_2\text{As}_2$	26.51(8)	0.393(1)	1.254(2)	32.3(2)	1.22	0.44	78
$\text{Sr}(\text{Co}_{0.85(2)}\text{Ni}_{0.15(2)})_2\text{As}_2$	25.1(3)	0.370(1)	1.217(1)	33.7(3)	1.32	0.39	70
$\text{Sr}(\text{Co}_{0.80(1)}\text{Ni}_{0.20(1)})_2\text{As}_2$	10.79(6)	0.328(2)	1.146(3)	24.2(4)	2.24	0.41	74
$\text{Sr}(\text{Co}_{0.75(1)}\text{Ni}_{0.25(1)})_2\text{As}_2$	3.08(1)	0.259(1)	1.018(2)	16.3(3)	5.29	0.60	107
$\text{Sr}(\text{Co}_{0.70(1)}\text{Ni}_{0.30(1)})_2\text{As}_2$		0.206(1)	0.907(2)	-0.15(1)			
$\text{Sr}(\text{Co}_{0.50(1)}\text{Ni}_{0.50(1)})_2\text{As}_2$		0.172(4)	0.828(1)	-84(3)			

TABLE IV: Parameters estimated from magnetization measurements of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ at $T = 2$ K. Spin-flop field H_{SF} , critical field H_c at which M_{ab} becomes approximately equal to M_c in Fig. 5 with increasing H , saturation moment μ_{sat} for $H \parallel ab$ and $H \parallel c$ obtained by extrapolating the high-field data in Fig. 5 to $H = 0$, angle-averaged saturation moments $\mu_{\text{sat,ave}}$ and $\mu_{\text{sat,c}}$, angle-averaged effective moment $\mu_{\text{eff,ave}}$ from Table II, and the Rhodes-Wohlfarth ratio p_c/p_{sat} where p_c is given by Eq. (9d) and $p_{\text{sat}} \equiv \mu_{\text{sat,ave}}/\mu_B$.

Compound	Field Direction	H_{SF} (T)	H_c (T)	μ_{sat} $\left(\frac{\mu_B}{\text{Co+Ni}}\right)$	$\mu_{\text{sat,ave}}$ $\left(\frac{\mu_B}{\text{Co+Ni}}\right)$	$\mu_{\text{eff,ave}}$ $\left(\frac{\mu_B}{\text{Co+Ni}}\right)$	p_c/p_{sat}
$\text{Sr}(\text{Co}_{0.987(1)}\text{Ni}_{0.013(1)})_2\text{As}_2$	$H \parallel ab$			0.037(3)	0.035(3)	1.33(1)	19(1)
	$H \parallel c$			0.0311(3)			
$\text{Sr}(\text{Co}_{0.96(1)}\text{Ni}_{0.04(1)})_2\text{As}_2$	$H \parallel ab$	0.66(5)	2.6(5)	0.067(4)	0.064(3)	1.340(4)	10.4(5)
	$H \parallel c$		3.4(3)	0.058(2)			
$\text{Sr}(\text{Co}_{0.94(1)}\text{Ni}_{0.06(1)})_2\text{As}_2$	$H \parallel ab$	0.91(4)	2.7(1)	0.118(1)	0.117(1)	1.259(2)	5.18(6)
	$H \parallel c$		4.3(2)	0.115(1)			
$\text{Sr}(\text{Co}_{0.90(1)}\text{Ni}_{0.10(1)})_2\text{As}_2$	$H \parallel ab$	0.49(2)	2.04(2)	0.166(3)	0.167(3)	1.254(2)	3.55(7)
	$H \parallel c$		3.37(5)	0.171(3)			
$\text{Sr}(\text{Co}_{0.85(2)}\text{Ni}_{0.15(2)})_2\text{As}_2$	$H \parallel ab$	0.41(2)	1.62(4)	0.165(1)	0.165(1)	1.217(1)	3.48(2)
	$H \parallel c$		2.91(3)	0.165(2)			
$\text{Sr}(\text{Co}_{0.80(1)}\text{Ni}_{0.20(1)})_2\text{As}_2$	$H \parallel ab$	0.23(3)	0.64(4)	0.128(2)	0.130(2)	1.146(3)	3.99(7)
	$H \parallel c$		1.3(2)	0.135(2)			
$\text{Sr}(\text{Co}_{0.75(1)}\text{Ni}_{0.25(1)})_2\text{As}_2$	$H \parallel ab$		0.60(6)	0.092(6)	0.094(5)	1.018(2)	4.5(2)
	$H \parallel c$		0.65(4)	0.098(3)			
$\text{Sr}(\text{Co}_{0.70(1)}\text{Ni}_{0.30(1)})_2\text{As}_2$	$H \parallel ab$		0.61(7)	0.048(4)	0.048(3)	0.907(2)	7.2(4)
	$H \parallel c$		0.58(8)	0.049(3)			

TABLE V: Spin-fluctuation parameters estimated with Takahashi's spin fluctuation theory in Eqs. (18b), (18c) and (21) from M^2 vs H/M and M^4 vs H/M isotherm measurements for $x = 0.04, 0.06, 0.10, 0.15, 0.20,$ and 0.30 . The fitted parameters are T_C (Curie-temperature), T_A (the width of the distribution of the dynamical susceptibility in the q space), and T_0 (the energy width of the dynamical spin fluctuation spectrum), and F_1 mode-mode coupling term. The error bars reflect systematic errors found from different field ranges of the fits.

Data Obtained from M^2 versus H/M Plots							
x	M_0 $\frac{\mu_B}{\text{Co+Ni}}$	slope ^a $10^{-8} \frac{[\mu_B/(\text{Co+Ni})]^3}{\text{Oe}}$	T_C (K)	T_{A1} (K)	T_{01} (K)	F_1/k_B (K)	T_C/T_{01}
0.04	0.0417(3)	2.67(2)	5	10874	783	40271	0.006
0.06	0.1075(1)	2.62(3)	20	10553	725	40954	0.028
0.10 ^b	0.1714(3)	3.75(1)		5583	290	28662	0.070
0.15 ^b	0.1694(2)	3.52(2)		5734	287	30549	0.070
0.20 ^b	0.1293(3)	2.38(2)		5886	205	45287	0.061
Data Obtained from M^4 versus H/M Plots at T_C							
x	M_0 $\frac{\mu_B}{(\text{Co+Ni})}$	slope $10^{-8} \frac{[\mu_B/(\text{Co+Ni})]^5}{\text{Oe}}$	T_C (K)	T_{A2} (K)	T_{02} (K)	F_1/k_B (K)	T_C/T_{02}
0.10	0.1666(2)	0.19(1)	20	3546	1343	2497	0.015
0.15	0.1664(1)	0.17(1)	20	3675	1214	2967	0.016
0.20	0.1255(2)	0.08(2)	12.5	3442	1226	2579	0.010

^aslope $\frac{M^2}{H/M}$ from M^2 vs H/M plots at $T = 2$ K

^bfrom $M(H)$ data at $T = 2$ K, T_C is obtained from M^4 vs H/M plots in the bottom part of the table

TABLE VI: Magnetic and spin fluctuation parameters for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ with $x = 0.04, 0.06, 0.10, 0.15, 0.20,$ and 0.30 . The spontaneous (saturation) moment per Bohr magneton $p_{\text{sat}} = M_{\text{sat},ab}/\mu_B$ ($M(H = 0 - 14$ T) at $T = 2$ K extrapolated to $H = 0$, from Fig 13), the effective magnetic moment μ_{eff} and Weiss temperature θ_p are obtained from magnetization measurements. The error bars reflect systematic errors found from different field range of the fits. p_c/p_{sat} is the Rhodes-Wholfarth ratio and $p_{\text{eff}}/p_{\text{sat}}$ is the Deuguchi-Takahashi ratio, where $p_{\text{eff}} = \mu_{\text{eff}}/\mu_B$ from Table III, and $p_c = \sqrt{1 + p_{\text{eff}}^2} - 1$ from Eq. (9d). For the ratio T_C/T_0 , the Curie temperatures T_C are given in Table V.

x	$p_{\text{sat}}\mu_B$ $\left(\frac{\mu_B}{\text{Co+Ni}}\right)$	$p_{\text{eff}}\mu_B$ $\left(\frac{\mu_B}{\text{Co+Ni}}\right)$	$p_c\mu_B$ $\left(\frac{\mu_B}{\text{Co+Ni}}\right)$	p_c/p_{sat}	$p_{\text{eff}}/p_{\text{sat}}$	θ_p (K)	T_C (K)	M^2 vs H/M	T_C/T_0	
									M^2 vs H/M	M^4 vs H/M
0.04	0.087(5)	1.340(4)	0.672(3)	7.7(4)	15.4(8)	8.9	5	0.006		
0.06	0.121(5)	1.259(2)	0.608(1)	5.0(2)	10.4(4)	21.1	20	0.028		
0.10	0.174(1)	1.245(2)	0.604(1)	3.43(2)	7.20(5)	32.3	20	0.070		0.015
0.15	0.173(2)	1.217(2)	0.575(1)	3.32(4)	7.03(8)	33.7	20	0.070		0.016
0.20	0.135(2)	1.146(3)	0.521(2)	3.85(7)	8.5(1)	24.2	12.5	0.061		0.010
0.25 ^a	0.094(5)	1.018(2)	0.427(1)	4.5(2)	10.8(5)	16.3				
0.30	0.057(3)	0.907(2)	0.350(1)	6.1(3)	15.9(8)					

^a p_{sat} from $M(H)$ at $T = 2$ K in Fig. 5, extrapolated to $H = 0$.

TABLE VII: Parameters γ , β and δ obtained by fitting the zero-field $C_p(T)$ data of $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ by Eq. (24). Also listed are the Debye temperature Θ_D calculated from β using Eq. (25) and the density of states at the Fermi energy $\mathcal{D}(E_F)$ calculated from γ using Eq. (27b). Another value of Θ_D is obtained by fitting $C_p(T)$ data by the Debye model according to Eq. (26).

Compound	γ (mJ/mol K ²)	β (mJ/mol K ⁴)	δ (10 ⁻³ mJ/mol K ⁶)	Θ_D (K)	Θ_D^a (K)	$\mathcal{D}_\gamma(E_F)$ $\left(\frac{\text{states}}{\text{eV f.u.}}\right)$
SrCo_2As_2	49.2(1)	0.291(5)	0.66(4)	322(1)	288(1)	20.86(4)
$\text{Sr}(\text{Co}_{0.96(1)}\text{Ni}_{0.04(1)})_2\text{As}_2$	53.66(8)	0.292(3)	1.52(3)	321(1)	299(2)	22.74(3)
$\text{Sr}(\text{Co}_{0.94(1)}\text{Ni}_{0.06(1)})_2\text{As}_2$	55.87(6)	0.414(2)	0.91(2)	286.3(4)	289(1)	23.68(2)
$\text{Sr}(\text{Co}_{0.90(1)}\text{Ni}_{0.10(1)})_2\text{As}_2$	57.35(7)	0.368(3)	0.732(3)	298(1)	300(1)	24.33(3)
$\text{Sr}(\text{Co}_{0.85(2)}\text{Ni}_{0.15(2)})_2\text{As}_2$	59.14(2)	0.368(1)	1.14(1)	297.8(2)	297(2)	25.07(1)
$\text{Sr}(\text{Co}_{0.50(1)}\text{Ni}_{0.50(1)})_2\text{As}_2$	42.82(9)	0.277(4)	4.25(5)	327(1)	285(4)	18.15(4)

^a Θ_D calculated from Eq. (26).

TABLE VIII: Parameters γ , β , δ , κ , and T_{SF} obtained by fitting the zero-field $C_p(T)/T$ data for $\text{Sr}(\text{Co}_{1-x}\text{Ni}_x)_2\text{As}_2$ below 10 K by Eq. (28). The values of Θ_{D} and $\mathcal{D}_\gamma(E_{\text{F}})$ for both spin directions were obtained from the β and γ values using Eqs. (25) and (27b), respectively.

Compound	γ (mJ/mol K ²)	β (mJ/mol K ⁴)	δ (mJ/mol K ⁶)	κ (mJ/mol K ²)	T_{SF} (K)	Θ_{D} (K)	$\mathcal{D}_\gamma(E_{\text{F}})$ ($\frac{\text{states}}{\text{eV f.u.}}$)
$\text{Sr}(\text{Co}_{0.80(1)}\text{Ni}_{0.20(1)})_2\text{As}_2$	58.9(2)	0.53(1)	0.00021(3)	-10.6(3)	10.5(2)	263(1)	25.0(1)
$\text{Sr}(\text{Co}_{0.75(1)}\text{Ni}_{0.25(1)})_2\text{As}_2$	60.5(1)	0.71(1)	0.00005(3)	-42.9(1)	5.39(4)	239(1)	25.7(1)
$\text{Sr}(\text{Co}_{0.70(1)}\text{Ni}_{0.30(1)})_2\text{As}_2$	56.4(1)	0.81(2)	0.0001(1)	-44.2(6)	4.89(5)	229(2)	23.9(1)

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