Cobalt monosilicide, CoSi, belongs to the new class of materials, featuring Weyl nodes in the bulk band structure, located very close to the Fermi energy. By shifting the chemical potential into these Weyl nodes using Fe-doping, quantum effects like the chiral anomaly and weak anti localization have been evidenced. However, the behavior of these effects with respect to the doping concentration is largely unexplored. Crystal structure and the electrical transport properties in single crystalline CoSi micro-ribbons with different Fe-doping are studied to identify the characteristic temperatures of electronic quantum and correlation effects. For the crystal structure analysis, transmission electron microscopy and powder X-ray diffraction analysis with a refined structural model is performed based on Le Bail fitting to analyze the lattice parameter. Hereby, the change of the lattice parameter of Co$_{1-x}$Fe$_x$Si is shown with increasing Fe content. In the electrical transport analysis, the occurrence of chiral anomaly and the weak anti localization effect in Fe-doped CoSi are presented. It is shown that these effect can be tailored by Fe-doping in CoSi.

1. Introduction

Ever since its discovery, a new class of fermionic materials fueled the curiosity of scientists. These materials are quantum materials with nontrivial band structure, a sub-class of the Weyl materials hosting Weyl fermions. Weyl fermions are topologically protected, highly stable quasiparticles with a chiral charge in bulk, which were first predicted by Hermann Weyl in 1929.[1] In contrast to classical Weyl semimetals, where the Weyl fermions carry a chiral charge of $\pm 1$, the Weyl fermions in these materials carry a chiral charge of up to $\pm 4$.[2] CoSi belongs to this new class of materials.[3] Recent studies on CoSi discovered helicoid-arc quantum states[4] and long Fermi arcs[5] that are a characteristic for Weyl materials and an experimental evidence of the nontrivial band structure.

This nontrivial band structure reflects in the transport properties of CoSi. CoSi crystallizes in the cubic B20 crystal structure (space group P2$_1$3 (198)) which has a broken space inversion symmetry. Bradlyn et al. predicted this space group P2$_1$3 (198) to host sixfold degenerated Weyl fermions.[2] Band structure calculations propose that three kinds of topological fermions exist in CoSi: those with a chiral charge of $\pm 1$, those with a spin 1 excitation with a chiral charge of $\pm 2$ and the so called Rarita–Schwinger–Weyl fermion (spin 3/2, chiral charge $\pm 4$).[3,6] The predicted Weyl nodes in CoSi were validated by angle resolved photo emission spectroscopy.[4] Band structure calculations showed two Weyl nodes located near the $\Gamma$ point. The first one is 20 meV above the chemical potential with a chiral charge of $\pm 4$. The second one is located 30 meV below the chemical potential with a chiral charge of $\pm 1$. The third one is 200 meV below the chemical potential at the R-point with a chiral charge of $\pm 4$.[6]

Since topologically protected quasi particles are massless, that is, feature a linear dispersion relation in the band structure, they have a considerable impact on the electrical transport properties. Thus, these states can be probed by electrical transport characterization, which proves to be a powerful method for in-depth investigation of Weyl materials. Characteristic quantum transport effects are, for example, quantum oscillations, large intrinsic anomalous Hall effect, weak anti-localization (WAL) and chiral anomaly. The WAL is caused by a phase
difference between the electron wave function and the time-inverse electron wave function leading to a sharp peak in the magneto-conductance. Chiral anomaly is caused by a broken chiral symmetry and leads to a positive contribution to the magneto-conductance for parallel configuration of the electrical and the magnetic field \( (E \parallel B) \). Weyl materials have also been reported to show distinct electron–phonon interactions as, for instance, the phonon-drag effect.\(^{[9]}\) The phonon drag effect is quite common for conductors, like copper, for example.\(^{[10]}\) Also, the formation of a charge density wave (CDW), that is, a static modulation of the conduction electrons to a correlated phase which is caused by a periodic structural distortion of the lattice, was theoretically predicted for some Weyl materials.\(^{[11–14]}\)

To probe the topological states by electrical transport, the chemical potential must be close to the Weyl nodes. Asanabe et al. observed a shifting of the chemical potential in CoSi by doping the composition with Fe in a range from 0% to 100%.\(^{[15,16]}\) In a preceded study on Fe-doped CoSi with 4% of Co replaced by Fe, we observed features of the chiral anomaly and WAl in combination with potential signatures of a CDW phase in the electrical transport.\(^{[17]}\)

Within this work, we exemplary evaluate crystal structure of Co\(_{1-x}\)Fe\(_{x}\)Si with from 0% to 20% Fe (0%, 4%, 10%, 20%). Additionally, we systematically characterized and analyzed the electrical transport properties of Co\(_{1-x}\)Fe\(_{x}\)Si compositions with 0% Fe, 2% Fe, 6% Fe, 10% Fe, and 20% Fe. To present a complete overview of the Co\(_{1-x}\)Fe\(_{x}\)Si compositions, we include the results of our previous study\(^{[17]}\) with 4% Fe in the presentation of the results.

### 2. Experimental Results

Poly-crystalline bulk samples of Co\(_{1-x}\)Fe\(_{x}\)Si with \( x = 0\% \), 2%, 6%, 10%, and 20% were synthesized by direct melting of the corresponding amounts of the components in a furnace with resistive heating followed by vacuum casting. Note that, the percent value gives the replaced Co by Fe in reference to the total amount of Co in the composition. The ingots were subsequently recrystallized by Bridgeman method. To verify the elemental composition (Table 1), inductively coupled plasma (ICP) and energy dispersive X-ray (EDX) during transmission electron microscopy (TEM) analysis was used. The crystal structure was analyzed via the TEM analysis. For in depth analysis of the crystal structure, X-ray diffraction (XRD) analysis in combination with a refined model based on the Le Bail (LB) method was used.

To investigate the electrical transport, we fabricated micro-ribbons by focused ion beam cutting to ensure single-crystallinity. Hence, the investigated samples were prepared out of one grain of the poly-crystalline bulk samples. The micro-ribbons have a defined dimension of 5 \( \mu \text{m} \times 60 \mu \text{m} \times 0.5 \mu \text{m} \) to ensure a homogeneous current flow. For the electrical characterization, the micro-ribbons were contacted by optical lithography and subsequent wire bonding to create a transport device, which was finally mounted to a quantum design Dynacool system in combination with external measurement devices (for details see Experimental Section). Following this preparation, detrimental influences by grain boundaries and current jetting could be suppressed, ensuring a homogeneous current flow over the characterized area (a photograph of the contacted ribbon is provided in Section S1, Supporting Information). Additionally, we analyzed the magnetic susceptibility in field cooled (FC) and zero field cooled (ZFC) mode with a magnetic properties measurement system (MPMS-XL7) with reciprocating sample option by quantum design.

### 3. Crystal Structure Analysis

#### 3.1. Transmission Electron Microscopy and Elemental Analysis

The crystal structure analysis was done by TEM on lamellae that were cut from the identical grain as the lamellae for the transport characterization. Thus, the chemical composition as well as crystalline structure obtained by the TEM study can be considered representative for the probed micro-ribbon. A compilation of this study is given in Figure 1. By EDX elemental analysis we determined the actual Fe content within the micro-ribbon that confirmed the expected composition. For the EDX analysis, 5 to 8 spots on each lamella were characterized and averaged. For in depth analysis of the elemental, composition we also performed ICP analysis using optical emission spectroscopy for the Fe containing compositions. For this, \( \approx 150 \) mg of the polycrystalline bulk samples was used and analyzed in three batches of \( \approx 50 \) mg. The ICP analysis was in good agreement with the initial composition used for the synthesis. The variation in the ICP analysis was below 0.5% for the different elements within one composition, pointing toward good

| Powder composition for the synthesis | Calculated composition | EDX results | ICP results |
|-------------------------------------|------------------------|-------------|------------|
|                                     | Co [at%] | Fe [at%] | Si [at%] | Co [at%] | Fe [at%] | Si [at%] | Co [at%] | Fe [at%] | Si [at%] |
| CoSi                               | 50       | 0        | 50       | 54       | 0        | 46       | 48.99    | 0.93     | 49.01     |
| Co\(_{0.92}\)Fe\(_{0.08}\)Si       | 49       | 1        | 50       | 50       | 1        | 49       | 47.08    | 2.74     | 48.89     |
| Co\(_{0.84}\)Fe\(_{0.16}\)Si       | 47       | 3        | 50       | 50       | 3        | 47       | 42.84    | 5.92     | 48.03     |
| Co\(_{0.74}\)Fe\(_{0.26}\)Si       | 45       | 5        | 50       | 45–48    | 5–10     | 46       | 39.68    | 9.94     | 49.10     |
| Co\(_{0.64}\)Fe\(_{0.36}\)Si       | 40       | 10       | 50       | 46       | 12       | 42       | 39.68    | 9.94     | 49.10     |
homogeneity within the samples. For the 2% and 6% Fe composition, only one batch of \( \approx 70 \) mg was characterized. The results of the ICP are included in Table 1.

Note that the nominal 10% Fe composition showed a slightly increased amount of Fe of 12% and lower Co and Si content. The variation of the composition was higher (±2%) between the different parts of the sample, too.

### 3.2. Powder X-Ray Diffraction and Refined Structural Model

For in depth analysis of the crystal structure, we used XRD analysis. The measured XRD patterns of the Co\(_{1-x}\)Fe\(_x\)Si powder samples are presented in Figure 2a. The phase analysis shows that all the observed reflections can be attributed to the CoSi structure (according to single-crystal data\(^{[18]}\) \( a = 4.4500(5) \) Å, space group \( P2_{1}3 \) (198) of the cubic syngony, powder diffraction file-2 [PDF-2] code 01-079-8014). With the increase of the nominal Fe content \( x \) from 0% to 20%, the Bragg angles of the reflections shift to smaller values reflecting an increase of the cubic unit cell parameter value \( a \).

One of the parameters important for the interpretation of the physical properties of the Co\(_{1-x}\)Fe\(_x\)Si solid solutions is the value of the Fe content \( x \) in them. Direct refinement of the Fe content \( x \) (in the case of the CoSi structure, this is the Co site occupancy coefficient \( p_{Fe/Co} \) with Fe atoms) and other structure parameters is in principle possible by Rietveld fitting\(^{[19]}\) the calculated XRD patterns to the measured ones.

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**Figure 1.** Scanning TEM images and electron diffraction pattern of each of the investigated compositions of Co\(_{1-x}\)Fe\(_x\)Si.

**Figure 2.** a) Measured XRD patterns of the Co\(_{1-x}\)Fe\(_x\)Si solid solution (\( x = 0\%\), 4\%, 10\%, and 20\%) powder samples. The inset illustrates the shift of reflection 2\( \Theta \) Bragg angles with an increase of Fe content \( x \) by the example of the reflection with \( hkl = 200 \). The XRD patterns are shifted with respect to each other for better visualization. b) LB fitting for Co\(_{1-x}\)Fe\(_x\)Si (\( x = 4\%\)) solid solution powder sample (reached agreement factors, weighted profile \( R_{wp} = 0.54\% \) and weighted profile corrected to background contribution \( cR_{wp} = 9.72\% \)). Miller indices \( hkl \) of the observed reflections are indicated.
However, the Rietveld structure refinement failed to obtain the Fe content with sufficient accuracy due to the close XRD scattering power of Co and Fe atoms.

There is another possibility to solve the problem of determining the composition of solid solutions, if an advantage will be taken, that the powder XRD allows to precisely determine the parameters of the unit cell of crystal phases, if the necessary angular corrections are introduced into the measured XRD pattern. In this case, taking into account that CoSi and FeSi are characterized by the same cubic structure, the Fe content \( x \) can be estimated from the value of the unit cell parameter \( a \) of the Co\(_{1-x}\)Fe\(_x\)Si solid solution according to Vegard's law.[20]

The simplest way to determine the parameters of a unit cell is by the Celsiz program[21] described in the Experimental Section, using only the values of the 2\( \Theta \) Bragg angles of the indexed observed reflections \( hkl \), corrected to zero shift and displacement. The results are presented in the Table 2. Another possibility to determine the precise unit cell parameters is to make a fit of the XRD pattern by the LB method,[22] which does not require knowledge of the structure, but only its symmetry, as it is described in the Experimental Section. For this analysis the program TOPAS[23] was used.

Analysis of the Williamson–Hall Plot (WHP)[24] and size strain plot (SSP)[25] evidences the absence of the microstrain in the powder crystallites. Taking into account the absence of the contribution of the microstrain broadening confirmed by WHP and SSP techniques, the LB fitting was carried out, refining the crystal size broadening only. Results of the LB fitting are summarized in the Table 2. An example of the LB fitting is presented in Figure 2b. The good quality of the fitting convincingly confirms single-phase state of all powder Co\(_{1-x}\)Fe\(_x\)Si samples.

As seen from the Table 2, all independent methods realized in the programs Celsiz, SizeCr, and TOPAS give the values of the crystallite size \( D \) and the cubic unit cell parameter \( a \) that match within their estimated standard deviations (e.s.d.s).

The values obtained by LB fitting are characterized by considerably smaller e.s.d.s, especially in case of the crystallite size \( D \).

Figure 3 presents the unit cell parameter \( a \) of Co\(_{1-x}\)Fe\(_x\) solid solution obtained by LB fitting in dependence on the nominal Fe content \( x \) together with the literature data from PDF-2 database (cards 01-079-8014, 01-089-7376, 00-038-1397, 01-079-0619, 01-086-0792, 01-088-1298, 01-089-2677 containing e.s.d. of the parameter \( a \) and having no status “deleted”[18-26-31] and all other cards 00-008-0362, 01-072-1328, 03-065-3296, 01-074-5667, 01-081-4605, 00-001-1271, 01-076-1748)[32-36] As is seen from Figure 3, the values of the unit cell parameter \( a \) from the Table 2, obtained in the current calculations, are well placed on a straight line, depending on the nominal \( x \). The linear dependence of the unit cell parameter \( a \) of a binary solid solution on the content of one of the elements indicates the validity of Vegard's law at least in the first approximation.

The coefficients of Vegard's law can be calculated from the exact values of the unit cell parameters of the extreme terms of the solid solution series, \( x = 0\% \) (CoSi) and \( x = 100\% \) (FeSi). However, the literature data show a rather large scattering of these parameters (Figure 3). Nevertheless, if the cards with the status “deleted” or without e.s.d.s will not be taken into account, the data scattering is significantly narrowed (Figure 3). In this case, only two points remain for the CoSi (\( x = 0\% \)) composition, one from the XRD powder data in this work, the second from the XRD single-crystal data from reference.[18] As the unit cell parameter of the CoSi compound (\( x = 0\% \)), the value \( a = 4.44448(2) \) Å from the Table 2 can be adopted, since the precision of the unit cell parameters are much higher from the powder XRD data than from single-crystal data (as opposed to atomic coordinates). As the parameter of the unit cell of the compound FeSi (\( x = 100\% \)), let take the average value of \( a = 4.4922(58) \) Å after averaging the data not canceled for this composition in Figure 3.

To double check the crystal structure, we also used the respective diffraction patterns for the investigated compositions. We identified the crystal structure in all different lamellae with the space group P2\(_1\)3 (198). Using the program ELDISCA,[37] we calculated the cubic unit cell parameter \( a \) for each composition as shown in Figure 3, using ten representative sets of data.

The resulting line graph of Vegard's law is shown in Figure 4. The values of the Fe content in the solid solutions under study, calculated from the unit cell parameters obtained in the LB fit, are plotted in the same Figure 4 and shown in Table 2.

As is seen in Table 2, the Fe content \( x \) calculated by means of Vegard's law from the unit cell parameters \( a \) of the solid solutions Co\(_{1-x}\)Fe\(_x\) obtained independently by program Celsiz least-square calculations and LB fitting matches well. The calculated Fe content \( x \) corresponds well to the nominal values of \( x \).
4. Electrical Transport Characterization

4.1. Room Temperature Hall Measurements and Temperature Dependent Resistivity

The resistivity at room temperature $\rho$, the Hall carrier density $n$, the carrier mobility $\mu$ and the temperature-dependent resistivity $\rho(T)$ are shown in Figure 5. The resistivity increases with increasing Fe content up to 20% Fe. Additionally, a local maximum was observed for 4% Fe. According to Pshenay–Severin et al., the addition of Fe to CoSi shifts the chemical potential closer to the Weyl point. The observed trend of a larger resistivity with increasing Fe content fits to a previous study of up to 4% Fe and was also observed by Asanabe et al. up to 10% Fe. Between 10% and 20% Fe, Asanabe et al. observed a decrease in $n$, contradicting the observation of this study.

Figure 5b shows $n$ at 300 K. The Hall effect sign indicates that electrons are the dominating carriers. By introducing 2% of Fe to CoSi, $n$ decreased from $7.2 \times 10^{21}$ to $7.2 \times 10^{20}$ cm$^{-3}$. With further increasing Fe content, $n$ increased again up to $5.6 \times 10^{23}$ cm$^{-3}$ at 20% Fe. In parallel, the carrier mobility ($\mu = (\rho n/e)$) increased for 2% Fe from 5 to 26 cm$^2$ V$^{-1}$ s$^{-1}$ and then decreased with increasing Fe. This behavior contradicts the report by Antonov et al., where a constant increase of the Hall resistance was observed with increasing Fe-content.

The density of states calculated by Imai et al. and later validated by Antonov et al. using density functional theory shows a minimum slightly above the Fermi energy for pure CoSi and an increasing density of states with decreasing Fermi energy. As $n$ is sensitive to the charge carrier concentration at the chemical potential, we can interpret that the chemical potential in the here synthesized CoSi lies slightly above the minimum represented in refs. [40] and [41]. By introducing Fe to the CoSi, the chemical potential is shifted downward through this minimum represented by a decrease followed by an increase in $n$ observed to higher Fe-content.

Note that the CoSi band structure in the vicinity of the Fermi energy consists of hole pockets around the $\Gamma$-point and electron valley at $R$-point of the Brillouin zone. The measured Hall signal of this multi-valley structure depends on the relative contributions, coming from the different valleys. Moreover, as it was shown in refs. [39, 41, 42] the inter-band energy dependent scattering plays an important role in electronic transport in CoSi and in Co-FeSi alloys. Thus, for a detailed Hall analysis, a multi-band model should be used. In our characterization, only linear behavior was observed at room temperature.

The relative change of $\rho$ with respect to the resistivity at 300 K was calculated by $\Delta \rho_{300K} = (\rho(T) - \rho(300K))/\rho(300K)$ and is shown in Figure 5c. The conduction mechanism of the different compositions show deviating trends and the shape of the individual curves indicates the occurrence of nontrivial transport mechanisms. The temperature dependent resistivity and its derivative is provided in Section S2, Supporting Information. We observed metallic-like conduction for 0%, 10%, and 20% Fe (decreasing $\rho(T)$ with decreasing T). In contrast, for 2% and 6% Fe and our previous study with 4% Fe, $\Delta \rho(T)$ develops a
local maximum located at increasingly higher temperature with increasing Fe. The maximum, observed in alloys with <6% Fe in this study, was also reported for 8% and 10% Fe above 300 K by Antonov et al.\(^{[46]}\) The general evolution of \(\rho(T)\) with both Fe-content and with temperature, is in qualitative agreement with previous observations by Asanabe.\(^{[15]}\) It is worth noting that a second increase in \(\Delta\rho(T)\) was observed between 40 and 2 K for the 2% Fe composition, and a change in the slope was observed in \(\rho\) between 75 and 40 K for the 6% Fe composition. This change was also observed and described in our previous study of the 4% Fe composition and was interpreted as potential signature of a CDW or spin density wave (SDW) phase occurring in the material.\(^{[27]}\) However, the analysis of the transport behavior has not yielded an unambiguous interpretation. For instance, comparable behavior was also observed in the spin gapless semiconductor Mn\(_2\)CoAl,\(^{[43]}\) metal–insulator–transition in ZrTe\(_5\),\(^{[44]}\) and also for an axionic CDW phase in (TaSe\(_4\))\(_2\)I.\(^{[45]}\)

### 4.2. Magnetization Characterization

The results of the magnetic characterization are shown in Figure 6. The temperature dependent magnetization was measured in ZFC and in FC mode. Hereby, the magnetic field for the characterization was either applied after cooling down the sample (ZFC) or before cooling down the sample (FC). The ZFC and FC magnetic susceptibility \(\chi(T)\) curves differ from one another for compositions 2%, 4%, and 6% Fe, where the ZFC \(\chi(T)\) curves show a local maximum with coincident temperature characteristics as the local maximum in \(\rho\) at that same compositions. For the pure CoSi and the 10% and 20% Fe compositions no noticeable difference was observed between the ZFC and the FC \(\chi\). The FC curves show paramagnetic behavior for all compositions Fe-containing samples. For pure CoSi, a transition from positive to negative \(\chi\) was observed at 50 K, indicating paramagnetic behavior below 50 K and diamagnetic behavior above. This magnetic transition, similarly reported earlier,\(^{[46]}\) was further validated by additional characterization at magnetic fields of 0.005 T and 0.02 T and in a vibrating sample magnetometer at 0.04 T (Section S3, Supporting Information). In general, an increase in \(\chi\) within three orders of magnitude was observed for the 0% composition to the 20% composition and increases with decreasing temperature. The fit accuracy of the SQUID data is provided in Section S4, Supporting Information. By doping CoSi with Fe, the chemical potential is shifted closer to the Weyl point (see subsequent data discussion). Thereby, an increase in the diamagnetic signal might be expected. This was not observed, most likely due to the nature of Fe-doping in CoSi. The substitutional incorporation of Fe-atoms seems to increase the paramagnetic signal rather than the diamagnetic one. This fits to the fact that FeSi is paramagnetic.\(^{[47]}\)

A similar behavior of the ZFC and FC \(\chi(T)\) as for the studied samples with 6% Fe and lower has been previously observed in other materials and was associated with spin waves in ferromagnets,\(^{[48]}\) SDWs in magnetic superconductors,\(^{[49]}\) magnetic nanoparticle systems,\(^{[50,51]}\) and metallic spin glass systems.\(^{[52]}\) In these cases, the temperature of the peak of the ZFC curve indicates the average energy threshold for either coherent magnetic ordering to take place, critical slowing down of superparamagnetic fluctuations, or freezing of magnetic moments into a frustrated metastable state mediated by RKKY-interaction.\(^{[53]}\) A maximum of the susceptibility is also observed in a number of other systems, such as nearly magnetic compounds with enhanced itinerant spin fluctuations\(^{[54]}\) or from magnetic Fe-nanoclusters in, for example, Fe\(_2\)O\(_3\)-nanoparticles in liquid solution\(^{[55]}\) and magnetic Fe-nanoclusters in bulk FeSi.\(^{[53]}\) Typically, these Fe-nanoclusters are in the size of a few atoms.\(^{[50]}\) Recently, it was shown by first principle calculations that local magnetic moments can induce spin-dependent scattering of both, surface and bulk electrons.\(^{[56]}\) Consequently, such Fe-nanoclusters may affect the local spin-configuration and with it also the magneto-resistance.

TEM investigations did not show the presence of metallic precipitates inside the samples. Note that Co and Fe are difficult to distinguish in TEM because of their neighboring position in the periodic table of elements. Thus, superparamagnetic effects cannot be excluded. Further studies of the frequency dependent susceptibility did not reveal indications of superparamagnetic behavior. Therefore, the interplay of spin textures predicted in CoSi,\(^{[6]}\) Fermi arcs, and the perturbation generated by the inclusion of small amounts of Fe on the spatial charge and spin distributions still needs to be elucidated.

### 5. Indications for WAL and Chiral Anomaly

#### 5.1. Magnetic Field Dependent Electrical Transport

In the following we analyze the series of Co\(_{1-x}\)Fe\(_x\)Si compositions with respect to the occurrence of WAL and chiral anomaly. For this, we characterized the magnetic field dependent electrical conductivity, in short magneto-conductivity, for all compounds in three different field directions. Hereby, the magnetic
Figure 7. The absolute change in the electrical conductivity ($\Delta \sigma = \sigma(B) - \sigma(0\,T)$) in dependence of the magnetic field. a,c,e,g,i) $\Delta \sigma$ in the $\mathbb{E}||\mathbb{B}$ and the $\mathbb{E} \perp \mathbb{B}$, b, d, f, h, j) $\mathbb{E} \perp \mathbb{B}$, oop configuration are shown at selected temperatures.

field was applied in parallel orientation to the electrical field ($\mathbb{E}||\mathbb{B}$) and in two perpendicular field orientations. For the first one, the magnetic field points perpendicular to the electrical field but lies in the sample surface plane (ip). This orientation is identified as $\mathbb{E} \perp \mathbb{B}$, oop. For the second one, the magnetic field is applied perpendicular to the electrical field and perpendicular to the sample surface plane (oop). This orientation is identified as $\mathbb{E} \perp \mathbb{B}$, oop.

The absolute change of the longitudinal electrical conductivity ($\Delta \sigma = \sigma(B) - \sigma(0\,T)$) for all compositions at selected temperatures in the magnetic field range from $-14$ to $14$ T is shown in Figure 7. We observed a qualitatively comparable shape in $\Delta \sigma$ for $\mathbb{E}||\mathbb{B}$, oop, since qualitatively $\mathbb{E} \perp \mathbb{B}$, ip and $\mathbb{E} \perp \mathbb{B}$, oop show comparable features. The complete characterizations are given in Sections S5–S9, Supporting Information. Figure 7a,c,e,g,i show $\Delta \sigma$ for $\mathbb{E}||\mathbb{B}$, Figure 7b,d,f,g,j display $\Delta \sigma$ for $\mathbb{E} \perp \mathbb{B}$, oop.

The largest change in $\Delta \sigma(B)$ of all the analyzed compositions in all orientations was found in the pure CoSi (0% Fe, Figure 7a,b). We observed a qualitatively comparable shape in the different orientations of $\mathbb{E}||\mathbb{B}$ and $\mathbb{E} \perp \mathbb{B}$. However, $\Delta \sigma(B)$ was about ten times larger for $\mathbb{E} \perp \mathbb{B}$ than for $\mathbb{E}||\mathbb{B}$. By doping 2% Fe (Figure 7c,d), we observed a sign change in $\Delta \sigma(B)$ for $\mathbb{E}||\mathbb{B}$. $\Delta \sigma(B)$ changed from $-11700$ S m$^{-1}$ for 0% to $4800$ S m$^{-1}$ for 2% Fe at 2 K and 14 T. $\Delta \sigma(B)$ for $\mathbb{E} \perp \mathbb{B}$ changed from $-132000$ S m$^{-1}$ for 0% to $-540$ S m$^{-1}$ for 2% Fe at 2 K and 14 T. A sharp peak developed below 10 K in each field orientation around zero field.

For the 6% Fe composition (Figure 7e,f), $\Delta \sigma(B)$ changes back to a negative value of $-5700$ S m$^{-1}$ for 6% at 2 K and 14 T. Nonetheless, a sign change was observed from negative to positive $\Delta \sigma(B)$ for temperatures above 23 K. For $\Delta \sigma(B)$ in $\mathbb{E} \perp \mathbb{B}$, we observed a $\Delta \sigma(B)$ of $-6300$ S m$^{-1}$ in $\mathbb{E} \perp \mathbb{B}$ at 2 K and 14 T. Here, a similar change in the sign of $\Delta \sigma$ above 25 K with a maximum positive $\Delta \sigma$ of 60 S m$^{-1}$ was found. Above 50 K, it returned to a negative value. Again, a peak developed below 10 K in each field orientation around zero field.

In the 10% Fe composition (Figure 7g,h), a negative $\Delta \sigma(B)$ in the whole temperature range was found independent on the magnetic field orientation. Additionally, the quantitative and qualitative $\Delta \sigma(B)$ in $\mathbb{E} \perp \mathbb{B}$ and $\mathbb{E}||\mathbb{B}$ differs only slightly, opposing the behavior observed for lower Fe content. Again, a peak developed below 10 K in each field orientation around zero field. In comparison to the 2% and the 6% Fe composition, an increase in $\Delta \sigma(B)$ was observed for $\mathbb{E} \perp \mathbb{B}$. In the 20% Fe composition (Figure 7i,j), we observed the same feature as in the 10% Fe composition. The maximum $\Delta \sigma(B)$ changed from $-15600$ S m$^{-1}$ for 20% to $-4200$ S m$^{-1}$ for 20% Fe composition at 2 K and 14 T.

5.2. Semiclassical Fitting of the Magnetic Field Dependent Electrical Transport

For the interpretation of the above-described trends in $\Delta \sigma(B)$, we used a semiclassical fitting model developed by Zhang et al. The model was designed for the electrical transport of Weyl materials in $\mathbb{E}||\mathbb{B}$ and includes the WAL and the chiral anomaly. In the Hakami–Larkin–Nagaoka equation, the WAL is described by a combination of the natural logarithm and the digamma function. In the approach by Lu et al., describing the WAL and the weak localization, the WAL is described by a sum over a combination of the digamma function and the natural logarithm. This approach was further developed to an integral over the digamma function in Weyl semimetals, which could be approximated by a $-\sqrt{B}$ dependence for high magnetic fields and a $-B^2$ dependence for low magnetic field. This part of the semiclassical fitting model has the scaling factor $C_{\text{WAL}}$. Zhang et al. further developed...
this semiclassical fitting including chiral anomaly. The chiral anomaly was described by a $B^2$ dependence scaling with $C_{\text{chiral}}$ and thus was added to the semiclassical WAL model.

This model was used in our previous study on the 4% Fe composition to separate the different transport signatures in $\Delta \sigma(B)$. The observed behavior of $C_{\text{pos}}$ in our data cannot be interpreted solely by the contributions of the chiral anomaly. The chiral anomaly is only present in $E//B$. For this, we introduce $C_{\text{pos}}$ as $C_{\text{chiral}} + C_{\text{CDW}}$ as further discussed in ref. [17]. Based on this, we analyzed $\Delta \sigma$ using Equation (1).

$$\Delta \sigma = C_{\text{pos}} \cdot B^2 - C_{\text{WAL}} \left( \sqrt{B^2 - B_0^2} + \frac{B^2}{B_0^2} + \gamma B^2 \frac{B_0^2}{B^2 + B_0^2} \right)$$

(1)

Here, $B_0$ is the critical field characterizing the transition between low and high magnetic field approximation. $\gamma$ is an additional scaling factor for the low magnetic field part. The results of this semiclassical fitting model are shown in Figure 8. $C_{\text{pos}}$ is shown in Figure 8a,c,e, while $C_{\text{WAL}}$ is shown in Figure 8b,d,f for $E//B$, $E.L.B$, $ip$ and $E.L.B$, $oop$, respectively. The fitting parameters from our previous study on the 4% Fe composition[17] are added to this figure. We will start the discussion with the $C_{\text{pos}}$ part.

We observed an increase in $C_{\text{pos}}$ for 2% Fe in $E//B$ at low temperatures. Below 10 K this increase was apparent for the other two directions, that is, $E,L,B$, $ip$ and $E,L,B$, $oop$. For 6% Fe, a contribution of $C_{\text{pos}}$ was found for temperature $>35$ K resulting in a peak between 35 K and 140 K but with notable contribution up to 300 K. A comparable peak was found for $E,L,B$, $ip$ (Figure 8c) and for $E,L,B$, $oop$ (Figure 8c), which is related to the change to a positive $\Delta \sigma$ shown in Figure 7f. For 0% Fe, 10% Fe, and 20% Fe no $C_{\text{pos}}$ contribution to $\Delta \sigma$ was observed.

A comparable peak of $C_{\text{pos}}$ in a shifted temperature range was also observed in our previous study of the 4% Fe composition (shown as reference in blue in Figure 8). In combination with $\rho(T)$, the peak was previously interpreted as the suppression of a CDW phase.[16] For the 6% Fe composition, we observed a similar feature $\rho(T)$ or more visible in the first derivative $d\rho/dT$ between 50 and 75 K (compare Section S2, Supporting Information). For the 2% Fe composition, we observed the same feature at 20 K, but only as a small change in the slope of $\rho(T)$.

For many of the characterized compositions and temperatures, $C_{\text{pos}}$ is close to zero. There is only a notable contribution from $C_{\text{pos}}$ for the samples containing 2%, 4%, and 6% Fe. They show a clear change in $C_{\text{pos}}$ in the corresponding temperature range (see Figure 8a,c,e). Hereby, $C_{\text{pos}}$ contributes in $E//B$ up to 300 K.

Comparing the values of $C_{\text{pos}}$, we observed an increase for the compositions with 2% and 6% Fe in different temperature ranges. No contribution of $C_{\text{pos}}$ was observed for the pure CoSi and the 10% and 20% Fe compositions. This reveals that the chemical potential was shifted all through the Weyl cone via doping with Fe.

We now continue with the discussion of $C_{\text{WAL}}$ (Figure 8b,d,f). The WAL is an effect independent on the field orientation.[60] In all characterized compositions containing Fe, we observed an contribution of $C_{\text{WAL}}$ below 15 K independent on the field orientation. However, there is a strong difference of the value of $C_{\text{WAL}}$ between the different compositions. For instance, the 10% Fe composition has the strongest $C_{\text{WAL}}$ contribution. For the 2% Fe and 4% Fe compositions the signature of the WAL is significantly smaller in comparison to the compositions with >6% Fe. For >6% Fe, the contribution of $C_{\text{WAL}}$ is comparable. For pure CoSi, $C_{\text{WAL}}$ did not contribute to $\Delta \sigma$. To emphasize this composition dependence, we plotted $C_{\text{pos}}$ and $C_{\text{WAL}}$ at 2 K versus the Fe content in Figure 9a,b.

To visualize the different overlapping effects in the different compositions, we created a sketch shown as inset in Figure 9a. We plotted the temperature ranges of the chiral anomaly, and the WAL with respect to the different compositions. The WAL

\[ \Delta \sigma = C_{\text{pos}} \cdot B^2 - C_{\text{WAL}} \left( \sqrt{B^2 - B_0^2} + \frac{B^2}{B_0^2} + \gamma B^2 \frac{B_0^2}{B^2 + B_0^2} \right) \]

Figure 8. Results of the semiclassical fitting for each field configuration. a,c,e) In the top row a,c,e) the parameter $C_{\text{pos}}$ is shown. b,d,f) In the bottom row we show the parameter representing the WAL contribution. A contribution was only observed below 15 K.
the Fe-content of the sample. We only observed a contribution of $C_{\text{pos}}$ for 2% to 6% Fe. $C_{\text{WAL}}$ increased with increasing Fe content. The inset in (a) visualizes the temperature windows of the observed effects chiral anomaly and WAL for the different Fe compositions.

was observed in all Fe containing compositions below 10 K, for 2% Fe below 15 K.

6. Conclusion

To emphasize the quality of the here synthesized Co$_{1-x}$Fe$_x$Si compounds we analyze the crystal structure and the elemental composition. The crystal structure was investigated using TEM and XRD characterization. The TEM analysis verifies large grains $>50 \mu m$ within the bulk samples. The EDX and ICP elemental analysis shows good agreement between the bulk samples and the micro ribbons. The refined analysis model for the XRD with LB fitting confirms Vegard's law of the compounds resulting in a continuous increase of the lattice parameter $a$ with increasing Fe content within the compound.

With the single crystalline micro ribbons, it is possible to tailor the presence of topological features in the transport properties by a shift of the chemical potential, which is realized here by doping pure CoSi with Fe-atoms on substitutional Co-lattice sites. The compositions with 2%, 4%, and 6% Fe show quite comparable properties with respect to the chiral anomaly and the WAL. The intended shift of the chemical potential close to the Weyl node was realized within these three compositions.

7. Experimental Section

Preparation of Single Crystalline Micro-Ribbons and Structural Analysis: Samples of Co$_{1-x}$Fe$_x$Si were prepared by direct melting of stoichiometric amounts of components in a furnace with resistive heating followed by vacuum casting. The ingots of cylindrical shape were recrystallized by Bridgeman method with inductive heating.

From these polycrystalline bulk samples, single crystalline micro-ribbons were prepared in the dimensions of about $50 \mu m \times 5 \mu m \times 300 nm$ using focused ion beam cutting (FEI Helios NanoLab 600i).

The crystal structure analysis was done by transmission electron microscopy (FEI Tecnai G2, 200kV acc. voltage, LaB$_6$ filament). Additionally, the EDX analysis and XRD analysis was performed. Results are shown in Table 1. The EDX analysis was done at the TEM lamellae at three to eight different spots. For the ICP analysis, 150 mg of one composition were taken and analyzed in three measurements to also have an overview on the homogeneity of the samples. Because of the small amount of material for the 2% and 6% composition, only one measurement was done here. The whole data is provided in the supporting online material in Table 1.

X-Ray Diffraction Analysis: For XRD measurements, the powder Co$_{1-x}$Fe$_x$Si (nominal $x = 0\%$, 4\%, 10\%, 20\%) solid solution samples were prepared on single-crystal background-free Si(111) sample holders. D2 PHASES powder X-ray diffractometer (Bruker AXS, Germany), in vertical Bragg–Brentano $\Theta$–$\Theta$ geometry, equipped with a position-sensitive semiconductor linear X-ray detector LYNXEYE (Bruker AXS, Germany), was utilized to measure the XRD powder patterns in the symmetric $\Theta$–$\Theta$ scan mode. X-ray tube with a copper anode was used to produce Cu-K$_{\alpha1,2}$ radiation, which was monochromatized by a Ni filter. During the measurements, the powder Co$_{1-x}$Fe$_x$Si samples were rotated around the axis of the sample holder to reduce the possible effects of the preferred orientation of the crystallites.

Additional measurements of the Co$_{1-x}$Fe$_x$Si powders mixed with certified standard Si640d silicon powder (NIST, USA) were performed to correct the detector zero shift ($\Delta \Theta_{\text{zero}}$) and the sample surface offset from the focus plane (displacement $\Delta \Theta_{\text{disp}}$) for the Co$_{1-x}$Fe$_x$Si sample XRD reflections utilizing the certified angle positions of the Si reflections as an internal standard.

The XRD patterns registered were analyzed by program EVA to obtain the Co$_{1-x}$Fe$_x$Si sample reflection parameters (Bragg angle values $2\Theta_{\text{hkl}}$, full width at half maximum, maximum ($I_{\text{max}}$), and integral ($I_{\text{int}}$) intensities) after introducing the necessary corrections to contribution of background and Cu-K$_{\alpha1,2}$ radiation.

The phase analysis and assignment of the Miller indices $hkl$ to the observed reflections were carried out using PDF-2 database.

The unit cell parameters were calculated by means of program Celis using the method of least-square fitting for the individual observed and calculated 2$\Theta$ values corresponding to every XRD reflection taken into account.

The obtained Co$_{1-x}$Fe$_x$Si reflection parameters were used in program SizeCr to determine the type of reflection profile and to construct the WHP and SSP graphs and estimate the mean values of the microstructure parameters, such as crystallite size $D$ and microstrain $\varepsilon$.\cite{24,25}

The unit cell and microstrain parameters were independently determined by the modeling and fitting of theoretical XRD patterns to experimental ones by the whole-pattern fitting LB methods.\cite{26} In contrast to the Rietveld structure refinement and fitting technique,\cite{19} the LB fitting does not need the structure model (coordinates of the atoms, atomic temperature factors, and occupancy coefficients of the atomic sites) and sample-related parameters, such as the parameters of the preferred orientation of the crystalline phase. As a result, the number of refined parameters in the LB fitting and possible correlations between them were significantly reduced in comparison to the Rietveld method. The only knowledge needed to start the LB fitting was the space group of the structure and rough values of the unit cell parameters. LB fitting method allowed the precise determination of the unit cell and microstructure parameters of the crystalline phases present in the sample.

Fabrication of the Electrical Transport Characterization Device: After transferring the micro-ribbon to a glass substrate, it was contacted by a lift-off process via laser lithography. For this, a bilayer resist system of LOR-3B and Ma-PI205 by micro-resist was used as positive resist. Before removing the resist, the electronic contacts were created by magnetron sputtering.
sputtering (5 nm of Cr as adhesion layer and 150 nm Au). The contact structure included several contacts for the electrical characterization, to check the measurement signal at different positions and choose the best signal to noise ratio.

Electrical Transport and Magnetic Characterization: For the electrical transport characterization, a 14T quantum design Dynacool system with rotation option was used at temperatures between 2 and 300 K. The electrical resistance was characterized using a sourcemeter Keithley 2400 and a nanovoltmeter Keithley 2182A. The accuracy of the Keithley 2401 sourcemeter is 0.027%+ 60 nA for the used current range of 1 mA. For the Keithley 2182A nanovoltmeter, the voltage was detected after it stabilized within 0.05% variation. Including the measurement errors of the geometrical dimensions into the calculation of error propagation, it resulted in an error of about <0.5 μΩm for Fe <4% and 0.5–1.5 μΩm for the other samples. Note that an inaccuracy in the determination of the geometrical dimensions shifts the complete temperature measurement curve quantitatively, but does not change the temperature dependent trends. The Hall characterization was performed on the poly crystalline bulk samples with direct Al-wedge bonding in Hall–Bar structure. Hereby, the sample thicknesses were in the order of 1–2 mm. For the characterization, the electrical transport option of the quantum design Dynacool system was used. The magnetic susceptibility was characterized at the bulk samples. Therefore, a quantum design 7T superconducting quantum interference device in a magnetic properties measurement system with reciprocating sample option was used. The sample was fixed between two straws before being mounted to the device.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

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