Superconductivity induced by ruthenium substitution in an iron arsenide: investigation of SrFe$_{2-x}$Ru$_x$As$_2$ ($0 \leq x \leq 2$)

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The magnetism in SrFe$_2$As$_2$ can be suppressed by electron doping through a small substitution of Fe by Co or Ni, giving way to superconductivity. We demonstrate that a massive substitution of Fe by isovalent ruthenium similarly suppresses the magnetic ordering in SrFe$_{2-x}$Ru$_x$As$_2$ and leads to bulk superconductivity for $0.6 \leq x \leq 0.8$. Magnetization, electrical resistivity, and specific heat data show $T_c$ up to $\approx 20$ K. Detailed structural investigations reveal a strong decrease of the lattice parameter ratio $c/a$ with increasing $x$. DFT band structure calculations are in line with the observation that the magnetic order in SrFe$_{2-x}$Ru$_x$As$_2$ is only destabilized for large $x$.

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I. INTRODUCTION

Soon after the discovery of superconductivity (SC) in doped FeAsO ($R =$ rare-earth element) materials\cite{Ishida07} investigations also focused on the structurally related AFe$_2$As$_2$ compounds ($A =$ alkaline, alkaline-earth, or rare-earth metal). In the latter structures, the Fe$_2$As$_2$ slabs are separated only by single elemental $A$ layers.\cite{Chen08} The compounds become superconductors if appropriately modified by substitutions on the $A$ site by alkali metal\cite{Chen08} or direct substitution within the Fe$_2$As$_2$ slab by Co\textsuperscript{6,7,8,9} or Ni.\textsuperscript{10} Recently, also the appearance of SC upon substitution of As by P was reported.\textsuperscript{11} Controlled tuning of the electronic structure by selective substitutions provides an opportunity to test and refine theoretical models since these substitutions can introduce charge carriers, modify the lattice parameters, and may significantly suppress the structural/magnetic transitions observed in the ternary parent compounds.\textsuperscript{2}

Application of external pressure has been understood as a “clean” alternative to substitutions in tuning the electronic state. AFe$_2$As$_2$ compounds indeed show crossover to SC at pressures as low as 0.4 GPa for CaFe$_2$As$_2$\textsuperscript{12,13,14} and high $T_c$ were observed (27 K at 3 GPa for SrFe$_2$As$_2$; 29 K at 3.5 GPa for BaFe$_2$As$_2$)\textsuperscript{15} Pressure reduces the antiferromagnetic (AFM) phase transition temperature ($T_0$) in SrFe$_2$As$_2$ and an abrupt loss of resistivity hints for the onset of SC\textsuperscript{16,17,18}. However, the nature of pressure (hydrostatic vs. anisotropic strains) in these experimental procedures is currently up for debate.\textsuperscript{12}

A feature common to both approaches (chemical substitution or external pressure) is the correlation of anisotropic changes in the crystal lattice with the suppression of the spin density wave (SDW) type of AFM. Transition-metal ($T$) substitution studies in AFe$_{2-x}T_x$As$_2$ carried out so far show a significant contraction of the tetragonal $c$ axis length\textsuperscript{6,7,10} and SC upon partial substitution of Fe by Co or Ni (electron doping) and an opposite trend if Fe is replaced by Mn (hole doping).\textsuperscript{20} For the latter substitution series the SDW transition temperature $T_0$ is not suppressed with increasing $x$ and no SC is observed.\textsuperscript{20} In contrast, in indirectly-doped Sr$_{1-x}$K$_x$Fe$_2$As$_2$ and Ba$_{1-x}$K$_x$Fe$_2$As$_2$\textsuperscript{20} the $a$ lattice parameter decreases with $x$ while $c$ increases, keeping the unit cell volume almost constant. For BaFe$_2$As$_{2-x}$P$_x$, an isovalent substitution where SC is observed for $x > 0.5$, both $a$ and $c$ decrease with $x$.\textsuperscript{20} The fact that substitutions within the Fe$_2$As$_2$ slab with other $d$-metals lead to superconductivity, albeit with lower $T_c$ than for indirect doping, favors an itinerant electronic theory, in contrast to the strongly correlated cuprates.\textsuperscript{20}

A point to note is that the modification of the electron count by substitutions is inevitably connected with structural changes in the Fe$_2$As$_2$ slabs. Unfortunately, for most substitution series only the unit cell dimensions are reported while further crystallographic data (bonding angles) are unknown. For pressure studies on Sr/BaFe$_2$As$_2$ compounds aiming at physical properties it is difficult to connect the results to variations of the crystal structure since corresponding compressibility studies are largely missing. Thus, currently, it is not known how the valence electron concentration and/or the structural parameters have to be modified by substitution in order to suppress the magnetism and eventually generate SC in AFe$_2$As$_2$ compounds.

Recently, Nath et al.\textsuperscript{22} reported on the physical properties of SrRu$_2$As$_2$ and BaRu$_2$As$_2$, which are isostuctural to SrFe$_2$As$_2$ and do not show SC.\textsuperscript{22} On the other hand, the isostuctural LaRu$_2$P$_2$ is a long-known superconductor with $T_c = 4.1$ K.\textsuperscript{22} In this communication we present results of a study of the solid solution SrFe$_{2-x}$Ru$_x$As$_2$. A massive substitution of Fe by nominally isoelectronic Ru suppresses the SDW-ordered state and bulk superconductivity is observed for $0.6 \leq x \leq 0.8$. Characterization of the SC state by magnetic susceptibility, electrical resistivity, and specific heat measurements shows a maximum $T_c$ of $\approx 20$ K. Detailed crystallographic information of all samples of the series are obtained from full profile refinement of powder X-ray diffraction data. Band structure calculations confirm that AFM order is only suppressed for large $x$, giving way to SC in SrFe$_{2-x}$Ru$_x$As$_2$.

In this study we obtain the pertinent crystallographic details for a complete substitution series with mag-
netically ordered, superconducting, and normal-metallic ground states. Such data are a prerequisite for a future comparative study of substitution systems (with or without superconducting phases) based on the SrFe$_2$As$_2$ parent compound. Another aspect is that substitution with nominally isovalent Ru – in contrast to the supposed electron doping by Co or Ni – is expected not to change the charge. In this way also no charge disorder is generated within the Fe$_2$As$_2$ slab. This is in contrast to substitutions with $d$ metals from the cobalt or nickel group. On the other hand, Ru substitution possibly will generate disorder due to its heavier mass and larger size. Both scattering mechanisms seem to limit the achievable $T_c$ of materials with in-plane substitutions, in contrast to SC material obtained through indirect doping of the Fe$_2$As$_2$ slabs by substitution on the A site.

II. EXPERIMENTAL AND CRYSTAL STRUCTURE

Samples were prepared by powder metallurgical techniques. Blended and compacted mixtures of precursor alloys SrAs, Fe$_2$As together with As and Ru powder were placed in glassy-carbon crucibles, welded into tantalum containers, and sealed into evacuated quartz tubes were placed in glassy-carbon crucibles, welded into tanneto substitutions with $d$ metals from the cobalt or nickel group. On the other hand, Ru substitution possibly will generate disorder due to its heavier mass and larger size. Both scattering mechanisms seem to limit the achievable $T_c$ of materials with in-plane substitutions, in contrast to SC material obtained through indirect doping of the Fe$_2$As$_2$ slabs by substitution on the A site.

The magnetic susceptibility was measured in a SQUID magnetometer (MPMS, Quantum Design). The electrical resistivity was determined by a four-point dc method (current density $<$ 3 A mm$^{-2}$). Due to the contact geometry the absolute resistivity could be determined only with an inaccuracy of $\pm$30%.

Band structure calculations were performed within the local density approximation (LDA) using the full potential local orbital code FPLO (v. 8.00) with a $k$-mesh of 24$\times$24$\times$24 $k$-points and the Perdew-Wang parameterization of the exchange-correlation potential. The used structural parameters were those from Table II.$^{24}$

III. RESULTS AND DISCUSSION

For all samples the crystal structure of ThCr$_2$Si$_2$ type (space group $I4/mmm$) was refined from powder XRD data by full profile methods (Sr in 2$a$ (0, 0, 0), Fe/Ru in 4$d$ (0, 1/2, 1/4), As in 4$e$ (0, 0, $z$)) (see Table I). In the powder X-ray diffractograms of samples with $x \geq 0.5$ broadening of (00$l$) reflections are observed suggesting some local disorder along [001]. Nevertheless, they give no evidence for superstructure formation due to long-range Ru ordering. The refined lattice parameters, the refined Ru occupancies, as well as EPMA unambiguously reveal the substitution of Fe by Ru. The nominal Ru contents are in good agreement with both the Ru occupancies from XRD and the EPMA data. The samples contained as minor impurity Ru$_{1-x}$Fe$_x$As. Upon exchange of Fe by Ru a strong linear decrease of the $c$ parameter of the unit cell is observed ($-9.7\%$ for $x = 2$). The tetragonal $a,b$ plane and with it the transition-metal distance $d_{T-M}$ expands by the substitution (by +6.2\% for $x = 2$). The increase of the distances $d_{T-M}$ with $x$ on the other hand is small (+1.1\% for $x = 2$) and the $z$ parameter of As decreases by only 0.25\%. Surprisingly, the exchange of Fe by the larger Ru atoms results only in a 2.0\% increase in the unit cell volume $V$. The by far largest structural effect of Ru (or Co) substitution is the strong decrease of the $c/a$ ratio, i.e. a strong strain-like deformation with respect to the crystal lattice of the ternary Fe parent compound. Correspondingly, the tetrahedral bonding angles $\alpha_{1,2}$ As–(Fe,Ru)–As depart from each other with increasing $x$. There are no visible discontinuous changes in these room-temperature structure data which could be connected to the various electronic ground state of SrFe$_{2-x}$Ru$_x$As$_2$.

![Image](image.png)

FIG. 1: (Color online) Magnetic susceptibility $\chi(T)$ of SrFe$_{2-x}$Ru$_x$As$_2$ samples in a magnetic field $\mu_0H = 2$ mT.

In Fig. 1 the low-field magnetic susceptibility of Sr(Fe$_{2-x}$Ru$_x$)As$_2$ samples is plotted. Weak diamagnetic signals for $T < 16$ K in warming after zero-field cooling (zfc) are already visible for a Ru concentration $x = 0.4$. However, the diamagnetic shielding signal increases dramatically from $x = 0.5$ to $x = 0.6$. For $x = 0.6, 0.7,$ and 0.8 the shielding comprises the full volume of the sample suggesting bulk SC. On the other hand, the Meissner effect (measured during field cooling) is extremely small. This peculiarity is also observed for other $A$(Fe$_{2-x}$Fe$_x$)$_2$As$_2$ materials and is probably due to strong pinning in materials with substitutions on the iron site, i.e. within the superconducting slab. The sample with $x = 1.0$ also does not show bulk SC but only a very small diamagnetism after zfc. The superconducting transition temperatures are listed in Table II however the transitions are rather broad.
TABLE I: Crystallographic and electronic data for SrFe$_{2-x}$Ru$_x$As$_2$: lattice parameters $a$, $c$, ratio $c/a$, cell volume $V$, refined (by WinCSD) positional parameter $z$ of As, interatomic distance $d_{T-As}$, refined Ru content $x$, intensity ($R_I$) and profile ($R_P$) residuals, SDW ($T_0$) and superconducting transition temperature ($T_c^\text{mag}$; crossing of the tangent to the upper part of the fc susceptibility transition with $\chi = 0$) and from specific heat ($T_c^\text{cal}$; from two fluid model fit, see text), idealized specific heat jump $\Delta c_p/T_c^\text{cal}$, linear term $\gamma$ to $c_p$. Debye temperature calculated from $\beta$. $T_c$ are only listed for bulk superconductors. fil. = observation of superconducting filaments.

| $x$ nom. | $a$ [Å] | $c$ [Å] | $c/a$ | $V$ [Å$^3$] | $z_{As}$ | $d_{T-As}$ [Å] | $x$ | $R_I/R_P$ ref. | $T_0$ [K] | $T_c^\text{mag}$ [K] | $T_c^\text{cal}$ [K] | $\Delta c_p/T_c^\text{cal}$ [mJ/mol K$^2$] | $\gamma$ | $\Theta_D$ [K]
|----------|--------|--------|-------|----------|--------|----------------|----|-------------|--------|----------------|----------------|---------------------|------|--------|
| 0.0$^a$  | 3.9243(1) | 12.3644(1) | 3.1507 | 190.5 | 0.3600(1) | 2.388(1) | 0 | - | 203 | - | - | - | - |
| 0.1      | 3.93210(3) | 12.3446(1) | 3.1347 | 190.9 | 0.3603(1) | 2.3911(3) | 0.12 | 3.7 | 6.3 | 190 | - | - | - |
| 0.2      | 3.94387(2) | 12.2905(1) | 3.1125 | 191.2 | 0.3602(1) | 2.3922(3) | 0.21 | 3.6 | 6.8 | 165 | - | - | - |
| 0.3      | 3.95145(3) | 12.2515(2) | 3.1014 | 191.4 | 0.3601(1) | 2.3924(3) | 0.31 | 3.8 | 10.3 | 140 | - | - | - |
| 0.4      | 3.96689(2) | 12.1772(1) | 3.0697 | 191.6 | 0.3599(1) | 2.3925(4) | 0.42 | 4.6 | 8.4 | 100 | fil. | - | - |
| 0.5      | 3.97720(4) | 12.1300(2) | 3.0499 | 191.9 | 0.3597(1) | 2.3927(5) | 0.50 | 5.3 | 14.1 | - | fil. | <2.0 | - |
| 0.6      | 3.99178(2) | 12.0635(1) | 3.0221 | 192.2 | 0.3599(1) | 2.3950(4) | 0.61 | 4.2 | 7.8 | - | 19.3 | 19.8 | 13.4 | 6.2 | 232 |
| 0.7      | 4.0057(2) | 12.0887(1) | 2.9983 | 192.6 | 0.3598(1) | 2.3973(5) | 0.99 | 3.3 | 6.3 | - | 19.3 | 20.1 | 11.6 | 7.3 | 229 |
| 0.8      | 4.01096(2) | 11.9835(1) | 2.9877 | 192.8 | 0.3598(1) | 2.3983(4) | 0.81 | 4.9 | 7.5 | - | 17.6 | 17.2 | 13.6 | 6.7 | 231 |
| 1.0      | 4.04437(6) | 11.8097(3) | 2.9200 | 193.2 | 0.3597(1) | 2.4055(5) | 1.03 | 4.5 | 10.2 | - | fil. | <2.0 | 0 | 6.9 | 243 |
| 1.5      | 4.09812(2) | 11.5301(1) | 2.8135 | 193.6 | 0.3593(1) | 2.4056(4) | 1.50 | 3.8 | 7.4 | - | - | - | - | - |
| 2.0$^{bc}$ | 4.16911(2) | 11.1706(1) | 2.6794 | 194.2 | 0.3591(1) | 2.4144(4) | 2 | 4.2 | 7.6 | - | - | - | - | 4.1$^b$ | 270$^b$ |

$^a$from Ref. 22
$^b_q = 4.1713$ Å, $c = 11.1845$ Å, $V = 194.6$ Å$^3$ (from Ref. 21)
$^c_q = 4.168$ Å, $c = 11.179$ Å, $V = 194.2$ Å$^3$ (from Ref. 22)

FIG. 2: (Color online) Corrected high-field magnetic susceptibility $\chi(T)$ of selected SrFe$_{2-x}$Ru$_x$As$_2$ samples.

This observation of diamagnetic traces and the appearance of superconducting filaments for low Ru concentrations is certainly due to microscopic inhomogeneities of the Ru distribution.

The high-temperature susceptibility (high-field data eventually corrected for small ferromagnetic impurities, Fig. 2) of SrFe$_{2-x}$Ru$_x$As$_2$ with $x \leq 1.5$ is generally paramagnetic and shows a typical linear increase with $T$ for $T > 100$ K, similar to that of compounds of the Co substituted system $\text{A}_2\text{As}_2$. The absolute values decrease systematically with the Ru content $x$ as does the linear $T$ dependence. SrRu$_2$As$_2$ finally is diamagnetic and shows no significant slope of $\chi(T)$ above 100 K, in agreement with Ref. [21]. No phase transitions (except for SC) are observed for $x > 0.5$. The samples with $x \leq 0.4$ display anomalies at around 200 K, 190 K, and 150–170 K for $x = 0, 0.1,$ and 0.2, respectively. These temperatures are close to the temperatures $T_0$ in Table I which were identified from the anomalies in the resistivity $\rho(T)$ (see below) which mark the SDW transition.

Due to the inaccuracy of the contact geometry we prefer to plot normalized electrical resistivity in Fig. 3. The room temperature resistivity values $\rho(300$ K) are 500–800 $\mu$Ω cm for samples with $x = 0$ and low Ru content $x$. The $\rho(300$ K) values decrease slightly with increasing $x$ and for SrRu$_2$As$_2$ we find $\rho(300$ K) of only $\approx 230$ $\mu$Ω cm. The temperature dependence of $\rho(T)/\rho(300$K) corroborates the superconducting transitions. Zero resistivity is observed for Ru concentrations of $x = 0.5$, 0.6, 0.7, and 0.8 at 7.0 K, 17.5 K, 16.0 K, and 18.0 K, respectively. The sample with $x = 1.0$ also shows a drop in $\rho(T)$ below 18 K, however $\rho = 0$ is not reached at 4 K, indicating only filamentary SC. Also, a small drop in $\rho(T)$ at low $T$ is already visible for $x = 0.4$.

The resistivity of the sample $x = 0.4$ however also shows a kink at $T_0 \approx 100$ K, the $x = 0.3$ sample at $\approx 140$ K. For lower Ru concentration $T_0$ increases continuously (see Table I). The kink in $\rho(T)$, which roughly coincides with anomalies in the high-field susceptibility (see above), is the signature of the SDW transition. 24,25,28
FIG. 3: (Color online) Electrical resistivity (normalized at 300 K) of SrFe$_{2-x}$Ru$_x$As$_2$ samples ($x$ as indicated on the curves). Data for $x = 0$ from Ref. [23].

FIG. 4: (Color online) Specific heat $c_p/T$ vs. $T^2$ of SrFe$_{2-x}$Ru$_x$As$_2$ ($x = 0.6, 0.7, 0.8, 1.0$). Data for zero magnetic field (red circles) are shown with the corresponding two-fluid (full black line) and BCS (orange dashed line) type fits (see text). For a field $\mu_0H = 9$ T only data (blue diamonds) are shown. Data for $x = 0.7, 0.8, 1.0$ are shifted by 0.04, 0.08, and 0.12 units upwards, respectively. Inset: difference of the specific heats ($\Delta c_p$) of the SC sample with $x = 0.8$ and the sample $x = 1.0$ (no bulk SC). Both the difference curves for zero and 9 T fields are given.

which is found at $T_0 = 203$ K in SrFe$_2$As$_2$ [23]. Interestingly, in Sr/BaFe$_{2-x}$Co$_x$As$_2$ crystals the resistivity for all $x > 0$ increases below $T_0$ while for unsubstituted Sr/BaFe$_2$As$_2$ $\rho(T)$ decreases below $T_0$ (see, e.g., Refs. [24]). In contrast, for SrFe$_{2-x}$Ru$_x$As$_2$ the resistivity decreases below $T_0$ for all concentrations of Ru. Generally, for a full opening of a gap due to an SDW ordering an increase of $\rho(T)$ would be expected. Instead, it seems that the behavior of $\rho(T)$ below $T_0$ is connected to the presence or absence of charge disorder caused by electron doping. Obviously, for SrFe$_{2-x}$Ru$_x$As$_2$ and undoped materials, in absence of such disorder the mechanism leading to a decrease of $\rho(T)$ below $T_0$ is dominating.

The specific heat of selected samples is given in Fig. [3]. Clear albeit broadened anomalies are seen close the transitions temperatures $T^{\text{max}}$ indicated by the low-field magnetization (see Table I). The sample with $x = 0.7$ shows a less pronounced transition than the samples with neighboring compositions. In agreement with the reduced resistive $T_c$ we conclude that this sample is inhomogeneous and of lower quality than the other samples. The sample with $x = 0.8$ displays a pronounced transition at 17.2 K. The composition SrFeRuAs$_2$ shows no bulk SC above 2 K. Since the specific heats of the two latter samples are very similar for $T > 20$ K the data for $x = 1$ may serve as a reference for phonon and normal electronic contributions to the $x = 0.8$ data. The inset in Fig. [3] shows the difference of the specific heat of the samples. The size of the resulting step $\Delta c_p/T_c$ as evaluated by the usual entropy-conserving construction (equal areas in $c_p/T$) for $H = 0$ is $18.4(1.8)$ mJ mol$^{-1}$ K$^{-2}$ and $T_c = 17.3(5)$ K.

In order to obtain further electronic and phononic properties from the specific heat, the data between 5.0 K and 25.5 K were fitted with a model including a phonon contribution (harmonic lattice approach $c_p = \beta T^3 + \delta T^5$) and an electronic term according to the weak coupling BCS theory or the phenomenological two-fluid model. The latter model is a good approximation for the thermodynamic properties of some strong coupling superconductors. Folding with a Gaussian simulates the broadening of the transitions due to chemical inhomogeneities. The parameters resulting from the fits are given in Table [I]. The relative specific heat step $\Delta c_p/T_c$ at $T^{\text{cal}}$ is quite similar for the three investigated SC samples. It has to be remarked that these values are smaller than the value obtained from the difference of the samples with $x = 0.8$ and $x = 1.0$. Nevertheless, the size of the specific heat step at $T_c$ is small and comparable to values observed for superconducting compositions of SrFe$_{2-x}$Co$_x$As$_2$ (10–13 mJ mol$^{-1}$ K$^{-2}$; hole doping within the Fe$_2$As$_2$ slab) but much smaller than for, e.g., Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ ($\sim 98$ mJ mol$^{-1}$ K$^{-2}$) [30]. The fits with the two-fluid models (shown in Fig. [3]) are superior to those with the BCS model. The ratio $\Delta c_p/(\gamma T^{\text{cal}})$ is $\approx 2.1$, significantly larger than the weak coupling BCS limit (1.43). Both these findings indicate strongly coupled SC in agreement with $\mu$SR measurements (see, e.g., Ref. [31]).

To understand the changes in the electronic structure upon substitution of Fe by Ru, we carried out band structure calculations for $x = 0, 0.5, 1, 2$. The partial Ru substitution was modeled by supercells. Since any SDW pattern would be strongly influenced by the choice of the particular supercell, especially for the larger Ru content, we decided to compare the stability with respect to mag-
netism applying the fixed spin moment (FSM) approach (cf. Fig. 3 in Ref. 28). The resulting curves are shown in Fig. 5. As one may expect, the magnetic moment and the energy gain due to magnetic ordering are reduced with increasing Ru content $x$. The destabilization is especially strong between $0.5 \leq x \leq 1$, the fully Ru substituted compound is clearly nonmagnetic. In accord with the classical Stoner picture for itinerant magnets, the suppression of magnetism originates from a strong decrease of the density of states (DOS) at the Fermi level $N(\varepsilon_F) = 4.9, 4.4, 3.5$ and $1.9$ states/(eV f.u.) for $x = 0, 0.5, 1, 2$, in agreement with experimental $\gamma$ values (Table 1).

In the real system, the magnetism will be further destabilized due to the Fe-Ru disorder. In contrast to the Co substituted compound, where the Co and Fe $3d$ states are almost indistinguishable (see inset of Fig. 5), the Ru $4d$ states differ considerably in the region close to $\varepsilon_F$. This difference has its origin in the different potential and lower site energy of Ru with respect to Fe. In turn, the distinct potential will be responsible for an enhanced scattering and therefore destabilize magnetic ordering. In the Co substituted compound, however, only the shift of the Fermi level due to the additional electron (see Fig. 5) leads to a nonmagnetic state.

Although our calculation illustrate semi-quantitatively the suppression of magnetism (clearing the scene for incipient SC) in SrFe$_{2-x}$Ru$_x$As$_2$, the real interplay between magnetism and the slight volume expansion for increasing Ru content, accompanied by a reduction of the $c$ axis and small changes of the As $z$ position is rather complex: the volume expansion leads to narrower Fe $3d$ bands that would stabilize magnetic order, but due to the $c$ axis contraction these states are simultaneously shifted to lower energy resulting in a reduced DOS at $\varepsilon_F$. However, the Fe $3d$ states are pushed up in energy upon substitution of Ru due to the lower site energy of the Ru $4d$ states, compensating the reduction of $N(\varepsilon_F)$ partially. Furthermore, the Stoner factor $I$ for Ru is significantly smaller than for Fe, disfavoring magnetic order further.

### IV. DISCUSSION AND CONCLUSIONS

To sum up, we have shown that the partial isovalent substitution of Ru for Fe in SrFe$_2$As$_2$ suppresses the SDW transition and gives rise to bulk superconductivity. Both end members of the series, SrFe$_2$As$_2$ as well as SrRu$_2$As$_2$, crystallize in the ThCr$_2$Si$_2$ type structure ($I4/mmm$) and there is no indication of a change or of a superstructure for intermediate Ru concentrations at the applied experimental conditions. This observation indicates that Fe and Ru are isovalent. The lattice parameters $a$ and $c$ vary linearly but in opposite directions with $x$. As seen from the ratio $c/a$ (decreases by 15 % for $x$ between 0 and 2) a strong anisotropic modification of the lattice (a compression of the tetragonal cell along $c$) is introduced by the exchange of Fe by Ru. While $z_{As}$ does not change significantly with $x$ this implies a large change of the two different As–Fe–As bonding angles. While for SrFe$_2$As$_2$ these angles are quite similar (110.50 and 108.90°) they deviate quite strongly in SrRu$_2$As$_2$ (119.43 and 104.73°). Bulk superconductivity exists in this series for a $c/a$ ratio (at room temperature) around 3. Electronic properties like the high-temperature susceptibility also show a smooth variation with $x$.

Whether any of these structural modifications is of special importance for the occurrence of superconductivity in SrFe$_{2-x}$T$_x$As$_2$-type substitution series remains to be determined. Currently, it can be stated that electron doping by Co (or Ni) is much more efficient in order to suppress the SDW and allowing for a superconducting ground state than a compression of the cell along $c$. The fact that Ru substitutions do not suppress the SDW state for as low concentrations as for Co, strongly indicates that Ru is isovalent to Fe in SrFe$_{2-x}$Ru$_x$As$_2$. In (optimally superconducting) SrFe$_{1.80}$Co$_{0.20}$As$_2$ the $c/a$ ratio is still quite large ($c/a = 3.132$), however 0.2 electrons were added per formula unit.

In order to compare all these parameters further detailed structural studies on various SrFe$_{2-x}$T$_x$As$_2$ systems are required. For the BaFe$_{2-x}$(Co,Ni,Cu)$_x$As$_2$ systems a first attempt in this direction was presented recently. Also, it is desirable to measure the compressibility of the compounds under hydrostatic pressure. Only such data would allow a comparison of pressure experiments with chemical substitutions studies. During revision of this work, superconductivity in several systems Sr/BaFe$_{2-x}$T$_x$As$_2$ with $T = Ru^{33,34}$, Rh$^{35}$, Pd$^{36}$, and Ir$^{37}$ was reported. For some of these systems the electronic state was already addressed by DFT methods. Experimentally, it appears that the critical temperatures are generally limited to $\approx 20$ K and that only for isovalent substitution (viz. Ru) the required level of substitution
is such high as reported here. All these new 3d, 4d, and 5d-metal substitutions deserve both an experimental as well as a sophisticated theoretical treatment, including the investigation of the influence of disorder and charge doping.

1. Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. 130, 3296 (2008).
2. M. Rotter, M. Tegel, D. Johrendt, I. Schellenberg, W. Hermes, and R. Pöttgen, Phys. Rev. B 78, 020503(R) (2008).
3. M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. 101, 107006 (2008).
4. K. Sasnal, B. Lv, B. Lorenz, A. M. Guloy, F. Chen, Y. Xue, and C. Chu, Phys. Rev. Lett. 101, 107007 (2008).
5. M. Rotter, M. Tegel, D. Johrendt, Phys. Rev. Lett. 101, 107006 (2008).
6. G. F. Chen, Z. Li, G. Li, W. Z. Hu, J. Dong, J. Zhou, X. D. Zhang, P. Zheng, N. L. Wang, and J. L. Luo, Chin. Phys. Lett. 25, 3403 (2008).
7. A. Leithe-Jasper, W. Schnelle, C. Geibel, and H. Rosner, Phys. Rev. Lett. 101, 207004 (2008).
8. A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, D. J. Singh, and D. Mandrus, Phys. Rev. Lett. 101, 117004 (2008).
9. G. Cao, C. Wang, Z. Zhua, S. Jiang, Y. Luo, S. C. Z. Ren, Q. Tao, Y. Wang, and Z. Xu (2008), arXiv:0807.1304v2.
10. N. Kumar, R. Nagalakshmi, R. Kulkarni, P. L. Paulose, A. K. Nigam, S. K. Dhar, and A. Thamizhavel (2008), arXiv:0810.0848v1.
11. L. J. Li, Q. B. Wang, Y. K. Luo, H. Chen, Q. Tao, Y. K. Li, X. Lin, M. He, Z. W. Zhu, H. G. Cao, and Z. A. Xu (2008), arXiv:0809.2009v1.
12. S. Jiang, C. Wang, Z. Ren, Y. Luo, G. Cao, and Z. Xu (2009), arXiv:0901.3227v1.
13. L. G. Akselrud, P. Y. Zavalii, Y. Grin, V. K. Pecharsky, B. Baumgartner, and E. Wölfel, Mater. Sci. Forum 133-136, 335 (1993).
14. M. Rotter, M. Tegel, V. Weiss, M. Schmitt, M. Hanfland, D. Kasinathan, U. Schwarz, R. Nath, Y. Singh, and D. C. Johnston (2009), arXiv:0901.4582.
15. M. Tegel, M. Rotter, V. Weiss, F. Ronning, and D. Johrendt, J. Phys.: Condens. Matter 20, 452201 (2008).
16. Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Phys.: Condens. Matter 20, 322204 (2008).
17. M. S. Torikachvili, S. L. Bud’ko, N. Ni, and P. C. Canfield, Phys. Rev. Lett. 101, 057006 (2008).
18. P. L. Allreza, Y. T. C. Ko, J. Gillett, C. M. Petrone, J. M. Cole, G. G. Lonzarich, and S. E. Sebastian, J. Phys.: Condens. Matter 21, 012208 (2008).
19. M. Kumar, M. Nicklas, A. Jesche, N. Caroca-Canales, M. Schmitt, M. Hanfland, D. Kasinathan, U. Schwarz, H. Rosner, and C. Geibel, Phys. Rev. B 78, 184516 (2008).
20. Y. Qi, L. Wang, Z. Gao, D. Wang, X. Zhang, and Y. Ma (2009), arXiv:0903.1270v2.
21. P. C. Canfield, S. L. Bud’ko, N. Ni, J. Q. Yan, and A. Kracher (2009), arXiv:0904.3134v1.
22. S. Paulraj, S. Sharma, A. Bharathi, A. T. Satya, S. Chandra, Y. Hariharan, and C. S. Sundar (2009), arXiv:0902.2728.
23. Y. Qi, L. Wang, Z. Gao, D. Wang, X. Zhang, and Y. Ma (2009), arXiv:0903.4967.
24. F. Han, X. Zhu, Y. Jia, L. Fang, P. Cheng, H. Luo, B. Shen, and H.-H. Wen (2009), arXiv:0902.1028v1.
25. L. Zhang and D. J. Singh (2009), arXiv:0904.3084v1.