Strain induced superconductivity in the parent compound BaFe$_2$As$_2$

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The discovery of superconductivity with a transition temperature, $T_c$, up to 65 K in single-layer FeSe (bulk $T_c = 8$ K) films grown on SrTiO$_3$ substrates has attracted special attention to Fe-based thin films. The high $T_c$ is a consequence of the combined effect of electron transfer from the oxygen-vacant substrate to the FeSe thin film and lattice tensile strain. Here we demonstrate the realization of superconductivity in the parent compound BaFe$_2$As$_2$ (no bulk $T_c$) just by tensile lattice strain without charge doping. We investigate the interplay between strain and superconductivity in epitaxial BaFe$_2$As$_2$ thin films on Fe-buffered MgAl$_2$O$_4$ single crystalline substrates. The strong interfacial bonding between Fe and the FeAs sublattice increases the Fe–Fe distance due to the lattice misfit, which leads to a suppression of the antiferromagnetic spin density wave and induces superconductivity with bulk $T_c \approx 10$ K. These results highlight the role of structural changes in controlling the phase diagram of Fe-based superconductors.
The discovery of Fe-based superconductors (FeSC) with critical temperatures, $T_c$, exceeding 50 K has stimulated many experimental and theoretical efforts to reveal the mechanism for superconductivity (SC) and to find compounds with even higher $T_c$. The band structure of the stoichiometric parent compounds yields two or three small hole pockets around $\Gamma = (0,0)$ and two electron pockets around $M = (\pi,\pi)$ in the folded zone. The nesting between electron and hole bands makes the commensurate spin density wave structure (SDW) with $Q = (\pi,\pi)$ plausible. In all FeSCs, the transition from the paramagnetic to the magnetic-ordered SDW phase is accompanied by the structural transition from the tetragonal $I4/mmm$ to the orthorhombic $Emmm$ phase. Doping due to injection of charge carriers (electrons or holes), chemical pressure and related disorder impairs the nesting conditions resulting in reduced structural transition temperature, $T_S$, and Neél temperature, $T_N$. At some finite doping, the magnetic phase gives room to SC. The vicinity to the SDW phase suggests remaining strong spin fluctuations are possibly responsible for the high-temperature SC. Therefore, the influence of doping on the phase diagram of FeSC is rather complex.

Another way to realize SC in FeSC is applying external pressure. Finite pressure suppresses the SDW and restores the tetragonal symmetry. As a result, SC occurs with $T_c$ comparable to that obtained by doping. However, the existence of SC, the value of maximum $T_c$ and the pressure under which SC occurs can vary considerably, depending on the level of hydrostaticity. By investigating the pressure diagram with different levels of hydrostaticity, it was found that a uniaxial component of the pressure dramatically affects the phase diagram of AEFe$_2$As$_2$ (AE=alkali earth element) compounds. It was suggested that the uniaxial pressure component is especially efficient for suppressing SDW order and responsible for a high $T_c$ of $\approx$ 35 K ($\approx$ 7–9). However, to the best of our knowledge, SC under uniaxial pressure has only been reported in CaFe$_2$As$_2$ with a folded zone of tetragonal symmetry. As a result, SC appears with $T_c$ of 65 K in single-layer FeSe can be induced by injection of charge carriers (electrons or holes), chemical pressure and related disorder. However, the influence of doping on the phase diagram of FeSe is rather complex.

In this article, we demonstrate SC in stoichiometric parent compound Ba$_{122}$ thin films solely by strain. We focus on epitaxial FeSe thin films, which are currently of great interest because of the ability to modify their electronic properties solely by change of the ambient pressure for stabilizing the SC state in FeSC.

In our experimental and theoretical efforts to reveal the mechanism responsible for controlling the phase diagram of isovalently Ru-doped Ba$_{122}$, we show that our data provide a deeper understanding of the mechanism responsible for controlling the phase diagram.

**Results**

**Structural analysis.** High-resolution reciprocal space maps (Fig. 1a) of the Ba$_{122}$ (1 0 9) and (1 0 11) diffraction peaks as well as the (2 0 6) peak of the MgAl$_2$O$_4$ (spinel) substrate reveal that the Ba$_{122}$ layers ($d \leq d_{sc}$, where $d_{sc}$ is the critical thickness of Ba$_{122}$) exhibit the same $a$ lattice parameter ($a = 0.404$ nm). Increasing the thickness of the Ba$_{122}$ layer to $d > d_{sc}$ leads to a relaxation of this layer, indicating a decrease of the $a$ lattice parameter (visualized by a shift of the Ba$_{122}$ diffraction peaks to larger $Q_x$ values). In the $0$–$2\theta$ scans (Fig. 1b), a clear trend is observed with increasing thickness, $d$, of Ba$_{122}$. The film with $d = 10$ nm ($S_{10}$) has a $c$ lattice parameter of 1.267 nm, which is reduced by about $3\%$ in comparison with the target value ($c = 1.301$ nm). For $d = 60$ nm, two different $c$ lattice parameters were found ($c_1 = 1.279$ and $c_2 = 1.301$ nm), which is explained by partial stress relaxation within the Ba$_{122}$ volume. Further increasing $d$ results in complete stress relaxation within the whole volume of the Ba$_{122}$ film. The whole set of lattice constants depending on the film thickness are summarized in Table 1.

The mechanism of stress relaxation is also seen in bright field micrographs of sample $S_{10}$ and $S_{60}$ taken by a transmission electron microscope (TEM). The images for the beam direction parallel to the [0 1 0] direction of the spinel substrate are shown in Fig. 1c,d. The thickness of the Fe and Ba$_{122}$ layers for $S_{10}$ is $\approx 27$ and 10 nm, respectively. The corresponding values for $S_{60}$ are 31 and 64 nm, respectively. In contrast to $S_{10}$, the sample $S_{60}$ contains cracks aligned perpendicular to the film surface of Ba$_{122}$ in most cases. Average distances between such cracks are in the range of a few $\mu$m and were also confirmed by atomic force microscopy (see Supplementary Fig. S2). Also, the root mean square roughness, $R_{\text{rms}}$, was determined to be 3.5 nm for sample $S_{60}$. This is much larger in comparison with the thinner samples ($R_{\text{rms},S_{10}} = 0.7$ nm, $R_{\text{rms},S_{60}} = 0.4$ nm). The observed cracks are the result of the relaxation process (shortening of the a axis of the Ba$_{122}$) when the critical thickness of the Ba$_{122}$ layer is reached. In the vicinity of the cracks, the Ba$_{122}$ layer is partially flaked from the Fe buffer layer. This leads to a formation of planar defects (probably several layers of BaO) parallel to the ab-plane and can be seen in Fig. 1d. For the films with $d > d_{sc}$, the observed relaxation process results in areas with properties similar to strain-free bulk Ba$_{122}$ and areas that are still strained by the underlying layers (see also the sketches in Fig. 2b). Further analysis by convergent beam electron diffraction indicates that the strained state in the Ba$_{122}$ layer is primarily determined by the Fe layer beneath. This is in accord with the previous observation of interfacial bonding between Fe and the FeAs sublattice of the Ba$_{122}$ (ref. 20).

**Magnetic properties.** The SC properties of the films were investigated using several complementary techniques. The temperature dependence of the dimensionless volume magnetic susceptibility $\chi_{\text{SC}}(T) = (m(T) - m(40\, \text{K}))/V_{\text{Ba}_{122}}H$ and the normalized resistance $R(T)/R(40\, \text{K})$ for films with various thickness are shown in Fig. 2, where $m(T)$ is the sample magnetic moment, $V_{\text{Ba}_{122}}$ is the volume of the Ba$_{122}$ layer and $H$ is the applied magnetic field. A decrease of $d$ results in an increase of the superconducting $T_c$ and the SC volume fraction of the films. The upturn of the susceptibility in the SC state is probably caused by the paramagnetic Meissner effect (Wohlfleben effect). The large value of the diamagnetic signal at low temperatures suggests that an essential volume of the films is in the SC state at low $T$ (see inset of Fig. 2a). The temperature dependence of the normal volume magnetic susceptibility $\chi_{\text{N}}(T) = (m(T) - m(300\, \text{K}))/V_{\text{Ba}_{122}}H$ for films $S_{60}$ and $S_{60}$ is shown in Fig. 2c. As can be seen, the amplitude of the SDW anomaly at $T_N \sim 130\, \text{K}$ is reduced in the thinner film ($S_{60}$), which is partially strained. Moreover, no signature of the SDW transition was observed for the films with $d < 60$ nm, suggesting...
Relaxation processes for sample S60 are clearly seen as a shift of the Ba122 reflections with regard to the substrate peak. The line is a guide for the eyes.

Table 1 | Structural and superconducting properties of the investigated samples.

| Sample name     | c-axis parameter (nm) | a-axis parameter (nm) | c/a | V = α²c (Å³) | Tc (K) | Thickness d (nm) |
|-----------------|-----------------------|-----------------------|-----|--------------|--------|-----------------|
| Ba122-target    | 1.3014 (1)            | 0.396                 | 3.29| 204 (1)      |        |                 |
| S10             | 1.302 (1)             | 0.396 (2)             | 3.29| 204 (2)      | 2      | 0               |
| S15             | 1.301 (1)             | 0.396 (5)             | 3.29| 204 (4)      | 24     | 0               |
| BaFe2As2 (ref. 25) | 1.3022 (2)            | 0.39633 (4)           | 3.29| 204.55 (4)  |        |                 |
| Ba2Fe14−xRuxAs2 (ref. 25) | 1.2749 (2)            | 0.40342 (5)           | 3.16| 207.49 (5)  |        |                 |

The critical temperatures (Tc) are evaluated from K(T) and χ(T). The Ba122 target is a polycrystalline sample. The reference samples are single crystals.

That in-plane tensile strain effectively suppresses long-range SDW order and SC does not coexist with SDW order in the thin films. Tc ≈ 140 K of the unstrained thick films is consistent with Tc ≈ 140 K observed in stoichiometric BaFe2As2 single crystals. Electrical transport properties. The SC transition width, ΔTc = Tc, onset – Tc, of the films is very large (Fig. 2b,d). For films with d < 30 nm, the decrease of resistance starts at relatively high temperatures Tc, onset = Tc, ill ≈ 35 K, whereas zero resistance is measured at Tc ≈ 10 K as shown in Fig. 2c,d. As can be seen in Fig. 2d, Tc, ill exactly corresponds to the temperature at which splitting between zero field-cooled and field-cooled branches of the susceptibility χ(T) curves is observed. In turn, a strong signal in susceptibility data for the thin films with d < 30 nm is observed below Tc only (Fig. 2a). From the ratio between diamagnetic moments at T > 10 K and in the low-T region, we concluded that in the range between Tc, ill and Tc ≈ 10 K, SC exists only in a small volume fraction of the films. This so-called filamentary SC7,26 (see Supplementary Fig. S3) is attributed to minor regions with slightly different strain state. The values of Tc and Tc, ill given in Table 1 and Fig. 4, are deduced from the method presented in Fig. 2d.

Critical current densities. The presence of the ferromagnetic Fe buffer layer and the extremely planar geometry of the films (large demagnetization factor) do not allow to estimate directly the SC volume fraction from the diamagnetic screening at low temperatures (see Fig. 2a). Therefore, we additionally measured the transport critical current density, Jc, to demonstrate that a large volume of the films is in the superconducting state below Tc. It is...
seen in Fig. 3a that self-field $J_c$ values in the strained film $S_{30}$ are comparable to the ones in optimally Co-doped Ba122 epitaxial thin films having a similar thickness $d \approx 30$ nm and $T_c \approx 22$ K (ref. 27). We suppose that the different $J_c$ and irreversibility field for sample $S_{30}$ and sample $S_{10}$ are due to different pinning properties rather than related to grain connectivity or other extrinsic factors. A dominant weak-link behaviour is excluded by the relative robustness of $J_c$ to external magnetic fields (see Fig. 3b, c). Moreover, we exclude a large variation of the cross-section for the superconducting currents as we observed a comparable diamagnetic signal for both films (see Fig. 2a). Thus, the relatively high $J_c$ values obtained from transport current measurements are an additional strong evidence for a large SC volume of the films with $d \leq 30$ nm.

**Discussion**

We suppose that the main mechanism of inducing SC and, therefore, controlling the phase diagram of Ba122 in our films is tensile in-plane strain. To investigate the possible role of charge transfer from the Fe buffer to the Ba122 layer similar to the case of FeSe single layers (see Introduction), we compared the strained films with a non-strained one grown on different substrates but having similar thickness of Ba122 and Fe-buffer layers. It was found that the unstrained films do not show any signature of a superconducting transition (see Supplementary Fig. S4). Thus, a sizable effect of the proximity to the metallic Fe buffer layer on $T_c$ is ruled out. We sum up our main findings in Fig. 4 and Table 1. As can be seen, the films with thickness $d < d_e \approx 30$ nm having a large SC volume fraction are completely strained. The Ba122 layer

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**Figure 2 | Magnetic susceptibility and resistance.** (a) Temperature dependence of the dimensionless magnetic susceptibility $\chi_{sc}(T) = (m(T) - m(40 \text{K})) / V_{Ba122}H$ in the superconducting state of the films with different thickness, where $m(T)$ is sample magnetic moment, $V_{Ba122}$ is the volume of the Ba122 layer and $H$ is the applied magnetic field (data are not corrected for demagnetization factors). The inset shows the region of bulk SC. (b) Temperature dependence of the normalized resistance $R(T)/R(40 \text{K})$ of the investigated thin films in zero magnetic field. (c) The normal state susceptibility $\chi_b(T) = (m(T) - m(300 \text{K}))/ V_{Ba122}H$. The amplitude of the SDW anomaly at $T_N \approx 130$ K is reduced in the thinner film ($S_{30}$), which is partially strained.

**Figure 3 | Critical current densities.** (a) Temperature dependence of self-field $J_c$ for sample $S_{30}$ and $S_{10}$ compared with data on Co-doped Ba122 taken from ref. 27. The relatively large $J_c$ values indicate bulk SC for the strained films. Bulk $T_{c,0}$ was taken for normalizing the data. The error bars are caused by the uncertainty in the film thickness due to the surface roughness (minimal and maximal size of Ba122 layer thickness). (b) and (c) provide $J_c(H)$ data for samples $S_{10}$ and $S_{30}$, respectively.
observation of filamentary SC at 35 K in regions with a different strain state suggests that $T_c \geq 35$ K is possible in Ba122 without doping or applying external pressure.

**Methods**

**Sample preparation.** The targets used for the thin film growth were prepared by a conventional solid state reaction process as described in refs 12,13,33.

The thin films were grown using pulsed laser deposition with a KrF excimer laser ($\lambda = 248$ nm). The preparation process took place in an ultra-high vacuum chamber with a base pressure of $10^{-9}$ mbar. Before the deposition, the substrate was heated up to 750°C to clean the surface. Subsequently, the Fe buffer layer of about 30 nm thickness was deposited at room temperature with a laser frequency of 5 Hz and then heated to 670°C to flatten the Fe surface30,32. This temperature was held for 30 min before the Ba122 layer was grown with a frequency of 10 Hz. The layer thickness was adjusted via the pulse number at constant laser energy and was confirmed by TEM for selected samples. To achieve homogeneous samples without thickness gradient, the substrate was rotated during the whole deposition.

**Structural characterization.** The $c$ lattice parameters were calculated from X-ray diffractiongrams (in a Bruker D8 Advance Diffractometer) in Bragg Brentano geometry using the Nelson Riley function, whereas the $a$ lattice parameters were taken out of the reciprocal space maps measured in a Panalytical X’pert Pro system.

TEM characterization of the samples was performed at FEI Tecnai-T20 TEM operating at an accelerating voltage of 200 kV. Preparation of the TEM lamellae was done by the focused ion beam technique (FEI Helios 600i) using a platinum protection layer and 3 kV accelerating voltage in the last focused ion beam preparation step.

Electron diffraction X-ray spectroscopy in TEM and auger electron spectroscopy were performed to check the Ba122 stoichiometry. Sensitivity factors for the auger electron spectroscopy spectra were evaluated using single crystals33. Parts of the thin film and the single crystal were sputtered, and the resulting spectra were analysed in a JEOL JAMP 9500F field emission Auger microprobe (see Supplementary Fig. S5). The resulting data are given in supplement and confirm that the thin films are stoichiometric.

**Transport and magnetic measurements.** Transport measurements were performed using a physical property measurement system (Quantum Design) with four-probe method. Magnetic measurements were performed in a Quantum Design direct current superconducting quantum interference device. In the normal state, electrical currents flow mainly through the metallic Fe buffer layer having a lower resistance than the Ba122 layer. Hence, it is not possible to measure the SDW/structural transition via transport measurements. For the measurements of the critical current density, $I_c$, the samples $S_{Ba}$ and $S_{Ru}$ were structured using ion beam etching with a stainless steel mask. Bridges of 0.45 mm width and 0.6 mm length were fabricated. $I_c$ was determined from $E(J)$ characteristics using the geometry parameters of the bridges and applying a 1 $\mu$V cm$^{-1}$ criterion ($J_{max}$ Lorentz force configuration).

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Finally, we conclude that the coherent interfacial bonding between Fe and the FeAs sublattice of the Ba122 results in an in-plane straining of Ba122 to the (001) plane of bcc Fe-buffer layer, that is, increasing $a$ and reducing $c$ lattice parameters of the Ba122 layer for thicknesses $d < d_c \approx 30$ nm. This suppresses magnetism and gives rise to bulk SC with $T_c \approx 10$ K. The large superconducting volume fraction of the films with $d < d_c$ is evidenced by a pronounced diamagnetic screening and by measurements of a finite transport critical current. The close similarities of crystal parameters and $T_c$ between thin films and Ru-doped Ba122 indicate that structural changes are one of the dominant factors for controlling $T_c$ of Ba122. Further, the

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**Author contributions**

J.E. fabricated samples, performed $R(T)$-, $AFM$-, $\theta$-20-, $I$- measurements, analysed the data, designed the experiments and prepared the manuscript. V.G. did magnetic characterization, analysed the data, designed the experiments and prepared manuscript. P.C. carried out the TEM characterization, M.H. performed the auger electron spectroscopy measurements and R.H. performed the measurements of reciprocal space maps. F.K. and K.I. fabricated Ba122-pulsed laser deposition targets for thin-film deposition and helped with preparation of the thin films. J.H. and D.V.E. prepared manuscript. B.H., L.S. and W.S. supervised the experiments and contributed to manuscript preparation. I.S. designed and directed the research. All authors discussed the results and implications and commented on the manuscript at all stages.

**Additional information**

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