Pressure-induced enhancement of the superconducting properties of single-crystalline FeTe$_{0.5}$Se$_{0.5}$

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Abstract
The pressure dependence, up to 11.3 kbar, of basic parameters of the superconducting state, such as the critical temperature ($T_c$), the lower and the upper critical fields, the coherence length, the penetration depth, and their anisotropy, was determined from magnetic measurements performed for two single-crystalline samples of FeTe$_{0.5}$Se$_{0.5}$. We have found pressure-induced enhancement of all of the superconducting state properties, which entails a growth of the density of superconducting carriers. However, we noticed a more pronounced pressure-induced enhancement of the superconducting properties of FeTe$_{0.5}$Se$_{0.5}$ than that in the critical temperature ($T_c$) with pressure. We have observed that the critical current density increases under pressure by at least one order of magnitude.

(Some figures may appear in colour only in the online journal)

1. Introduction

The discovery of superconductivity in the Fe-based oxypnictide compounds [1] has sparked tremendous interest and opened up new perspectives in the field of superconductivity [2–6]. To date, the following groups of Fe-based superconductors are known: REOFeAs (‘1111’, RE = rare earth) [1], AFe$_2$As$_2$ (‘122’, A = alkaline earth) [2], LiFeAs (‘111’) [3], Fe(Se,Ch) (‘11’, Ch = S, Te) [4, 5], and Sr$_2$Mo$_2$FePn (‘21311’, M = Sc, V, Cr, and Pn = pnictogen) [6]. Within the ‘11’ group, pure FeSe exhibits superconductivity below $T_c \approx 8$ K [4]. The tetragonal compounds FeSe and FeTe$_{1-x}$Se$_x$ have quite a simple structure, with Fe and Te/Se layers additionally with Fe excess, alternating along the c-axis [4, 7, 8]. These compounds have attracted much interest because of their similarities to the iron–selenide family A$_x$Fe$_{2-y}$Se$_2$ by intercalating alkaline earth atoms (A = K, Rb, Cs) between the FeSe layers [14–16]. However, $T_c$ is found to decrease with pressure and is fully suppressed at 90 kbar for K$_x$Fe$_{2-y}$Se$_2$ [17] and at 80 kbar for Cs$_x$Fe$_{2-y}$Se$_2$ [18]. The critical temperature is very weakly dependent on pressure below 10 kbar, suggesting that $T_c$ is almost independent of small variations of the lattice constants.

In the case of FeTe$_{0.5}$Se$_{0.5}$, the $T_c$ increases with $P$ [19–21] up to 26.2 K at 89 kbar [11]. Interestingly, a similar high $T_c \approx 30$ K is attained in the iron–selenide family A$_x$Fe$_{2-y}$Se$_2$ by intercalating alkaline earth atoms (A = K, Rb, Cs) between the FeSe layers [14–16]. However, $T_c$ is found to decrease with pressure and is fully suppressed at 90 kbar for K$_x$Fe$_{2-y}$Se$_2$ [17] and at 80 kbar for Cs$_x$Fe$_{2-y}$Se$_2$ [18]. The critical temperature is very weakly dependent on pressure below 10 kbar, suggesting that $T_c$ is almost independent of small variations of the lattice constants.

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tetrahedra, noticed at 110 kbar in x-ray diffraction studies at room temperature [20, 22].

However, pressure studies of superconductivity in Fe(Se, Ch) system have been limited mainly to a tuning of \( T_c \). The pressure dependence of the upper critical field \( (H_{c2}) \) has been investigated for polycrystalline FeSe only [10]. Still, nothing is known about the pressure dependence of the lower \( (H_{c1}) \) and the upper critical fields and their anisotropies for single-crystalline FeTe\(_{1-x}\)Se\(_x\). There is a lack of data on the pressure impact on the critical current density \( (j_c) \) in FeTe\(_{1-x}\)Se\(_x\). Since we have established earlier [23] that the sharpness of the transition to the superconducting state in FeTe\(_{1-x}\)Se\(_x\) is evidently inversely correlated with the crystallographic quality of the crystals, we decided to perform pressure studies of two FeTe\(_{0.5}\)Se\(_{0.5}\) single crystals of significantly different crystallographic qualities.

The lower critical field, related to the London penetration depth, provides information about the density of superconducting carriers. The upper critical field, directly related to the coherence length, and its temperature dependence, provide some information about the pairing mechanism and pairing strength. Both microscopic quantities, together with the critical current density, are important for application purposes as well. In this paper, the pressure dependence of the lower and the upper critical fields and of the critical current density in FeTe\(_{0.5}\)Se\(_{0.5}\) is presented. The hydrostatic external pressure, up to 11.3 kbar, has led to a more pronounced increase in superconducting carrier density than that in the critical temperature, what may indicate the appearance of a mechanism limiting the increase of \( T_c \) with pressure.

2. Synthesis and experimental techniques

Single crystals of nominal composition FeTe\(_{0.5}\)Se\(_{0.5}\) were grown using Bridgman’s method. The studied samples were prepared from stoichiometric quantities of iron chips (3N5), tellurium powder (4N), and selenium powder (pure). All of the materials were weighed, mixed and stored in a glove box in argon atmosphere. A double walled evacuated and sealed quartz ampule with the starting materials was placed in a furnace with a vertical gradient of temperature equal to \( \sim 1.2 ^\circ C \) mm\(^{-1}\) for sample I and \( \sim 0.6 ^\circ C \) mm\(^{-1}\) for sample II. The material was synthesized for 3 h at a temperature of 730°C and then the temperature was increased up to 920°C. After melting, the temperature was held for 3 h, then the sample was cooled down to 500°C at a rate of 1.5°C h\(^{-1}\) (sample I) or 3°C h\(^{-1}\) (sample II) and then to 200°C at a rate of 60°C h\(^{-1}\) for both samples, and finally cooled down with the furnace to room temperature. As a result, we obtained two single crystals with different crystallographic qualities. In our case, the crystal quality was determined by the \( \Delta \omega \) value, describing the full width at half maximum (FWHM) of the 004 x-ray diffraction peak, obtained in \( \omega \) scan measurements, since changes in the c-axis lattice constant are very sensitive to the variation in chemical composition of the studied materials [23]. The 004 peak is relatively intense and appears at sufficiently large angles to get a good angular resolution. The crystals, with \( \Delta \omega \) values equal to 10.32 (labeled as sample I) and 16.65 (labeled as sample II) arc min, were grown with velocities of \( \sim 1.2 \) and \( \sim 5.2 \) mm h\(^{-1}\), respectively. The obtained single crystals exhibited a (001) cleavage plane and sample I with better crystallographic quality also had well developed (100) natural planes.

The quantitative point analysis on the cleavage plane of the crystals was performed by field emission scanning electron microscopy (FESEM) with a JEOL JSM 7600F operating at 20 kV incident energy coupled with Oxford INCA energy dispersive x-ray spectroscopy (EDX). The average chemical compositions of the crystal matrices checked by scanning electron microscopy (SEM) and EDX analysis (accuracy \( \pm 0.02 \)) were Fe\(_{1.00}\)Te\(_{0.58}\)Se\(_{0.42}\) and Fe\(_{1.01}\)Te\(_{0.57}\)Se\(_{0.43}\) for samples I and II, respectively.

The magnetic measurements were carried out on single-crystalline samples of roughly rectangular shape, in the temperature range of 2–300 K, with a magnetic field of up to 50 kOe, using a Quantum Design superconducting quantum interference device magnetometer. The magnetic field was applied parallel to the c-axis of the crystal and to the ab (001) plane, which is perpendicular to the c-axis. A hydrostatic external pressure up to 11.3 kbar was applied, using an easyLab Technologies Mceell 10 pressure cell with Daphne 7373 oil [24], this being considered as the best pressure medium from the point of view of the smallest decrease of pressure with decreasing temperature, at least in the pressure range above 7 kbar [25]. High-purity Sn wire (0.25 mm in diameter) was employed as an in situ manometer. The background signal associated with the pressure cell was subtracted on the basis of results obtained under ambient pressure for the sample placed in the pressure cell and for the sample without the pressure cell. We noted that the background contribution does not influence the obtained results. The measurements of ac susceptibility (field amplitude 1 Oe, frequency 10 kHz) were performed with a Quantum Design physical property measurement system (PPMS).

3. Results and discussion

3.1. The critical temperature

For single crystals of FeTe\(_{0.5}\)Se\(_{0.5}\), noticeable differences between the initial chemical composition and that estimated by EDX as well as a significant difference in the FWHM of the 004 x-ray diffraction peak \( (\Delta \omega) \) are visible (see, for example, [23]). Usually, they are attributed to a separation of phases with different Se/Te ratios, as reported in several papers [9, 26, 27]. However, the data obtained for monophase single crystals of FeTe\(_{0.65}\)Se\(_{0.35}\) [23] indicated that the narrowest transition to the superconducting state (width \( \sim 0.6 \) K) is exhibited by single crystals with relatively large values of \( \Delta \omega \) equal to 6 arc min. Furthermore, the decrease in the \( \Delta \omega \) value was found to be correlated with the increase of the width of the transition (90%–10% criterion). This correlation suggests that the disorder in some sense enhances the superconductivity in the FeTe\(_{1-x}\)Se\(_x\) system, and the
properties of the superconducting state of FeTe_{1-x}Se_x are very sensitive to the defects present in the sample [23].

The main aim of our work was to study the pressure effect on the intrinsic superconducting properties of FeTe_{0.5}Se_{0.5}. Since noticeable differences in the crystallographic quality were found among the crystals grown in various conditions, we decided to perform all of the measurements for two crystals of significantly different crystallographic qualities, i.e., for samples I and II with different Δω values for the 004 x-ray diffraction peak.

Figure 1(a) shows the temperature dependences of the real (4πχ′') and imaginary (4πχ′′) parts of the ac magnetic susceptibility measured in 1 Oe of ac field with 10 kHz in warming mode for two single crystals, grown with different cooling velocities and vertical gradients of temperature. The presented data were normalized to the ideal value of −1 for the real part of the ac susceptibility for better comparison of the susceptibility data obtained for the samples with different shapes and therefore subjected to different demagnetizing fields. The inset in the lower panel of figure 1(a) shows the variation of 4πχ′ for the crystals in the vicinity of T_c. It is obvious that, despite the significant difference in the width of the transition to the superconducting state, both of the crystals with different crystallographic quality are characterized by almost identical onsets of T_c.

Figure 1(b) presents the temperature dependence of the dc magnetization measured for sample I in a wide temperature range up to 300 K in zero-field cooling (ZFC) and field-cooled warming (FCW) modes in a magnetic field of 10 kOe, applied parallel to the c-axis of the studied single crystal under ambient pressure and at a hydrostatic pressure of 10.4 kbar. Similar behavior—not shown—was found for sample II. In the presented data, there is a clearly visible transition at about 130 K, most likely related to spin reorientation of the Fe_7Se_8-type minor phase or to a Verwey transition in FeO [28], coexisting in the crystal with the major tetragonal phase of FeTe_{0.5}Se_{0.5} [29, 30]. The magnetization does not exceed 1.9 emu g⁻¹, therefore the volume fraction of Fe_7Se_8-type phase or of FeO should not be greater than a few per cent. Importantly, both temperature dependences of magnetization, at ambient and at hydrostatic pressure, are characterized by almost identical shapes, indicating an absence of structural transition under pressure.

The temperature dependences of the magnetic susceptibility in the vicinity of T_c for H ∥ c-axis (upper panel) and H ∥ ab-plane (lower panel), measured under ambient pressure and applied hydrostatic pressure up to 10.4 kbar in a dc field of 10 Oe for sample I are presented in figure 2(a). The critical temperature was defined as the point on the x-axis where the M_{ZFC}(T) curve deviates from a constant, temperature independent background value. The almost linear dependence of the ZFC magnetic susceptibility below T_c, approximated well by parallel lines shifted to lower temperature with increasing pressure, indicates that the superconducting transition width is almost unaffected by pressure, at least in the studied, relatively narrow, pressure range. A significant divergence between the M_{ZFC} and M_{FCW} curves indicates relatively strong pinning of vortices for both H ∥ c-axis and H ∥ ab-plane even for the sample of better crystallographic quality (figure 2(a)). It was found that T_c increases linearly with pressure in the investigated pressure range from about 14 K at ambient pressure up to about 21 K at P = 10.4 kbar, for both H ∥ c-axis and H ∥ ab-plane (upper panel of figure 2(b)). This confirms earlier reports on T_c increase, for the FeTe_{0.5}Se_{0.5} compound, in the pressure range of 0–10 kbar [19–21]. The T_c(P) dependence for sample I, given in the upper panel of figure 2(b) by the thick solid line with the pressure coefficient dT_c/dP = 0.67(5) K kbar⁻¹, is a result of fitting of the linear dependence with the least...
Figure 2. (a) Temperature dependences of the dc magnetic susceptibility in the vicinity of the critical temperature for \( H \parallel c\)-axis (upper panel) and \( H \parallel ab\)-plane (lower panel), measured under ambient pressure and applied hydrostatic pressure up to 10.4 kbar in a dc field of 10 Oe for sample I. (b) The pressure dependence of the critical temperature determined for both \( H \parallel c\)-axis and \( H \parallel ab\)-plane magnetic field configurations for sample I (upper panel) and sample II (lower panel). The \( T_c(P) \) dependencies given by the solid lines are the results of fitting of the linear dependence with the least square method.

The temperature dependence of the dc magnetic susceptibility in the vicinity of the critical temperature for \( H \parallel c\)-axis (upper panel) and \( H \parallel ab\)-plane (lower panel), measured under ambient pressure and applied hydrostatic pressure up to 10.4 kbar in a dc field of 10 Oe for sample I. (b) The pressure dependence of the critical temperature determined for both \( H \parallel c\)-axis and \( H \parallel ab\)-plane magnetic field configurations for sample I (upper panel) and sample II (lower panel). The \( T_c(P) \) dependencies given by the solid lines are the results of fitting of the linear dependence with the least square method.

3.2. The thermodynamic parameters—the upper and the lower critical fields

In order to estimate the change in the anisotropic thermodynamic parameters of a single crystal of FeTe\(_{0.55}\)Se\(_{0.5}\) subjected to hydrostatic pressure, we have evaluated the temperature dependences of the upper and the lower critical fields in two geometries, \( H \parallel c\)-axis and \( H \parallel ab\)-plane (up to 50 kOe), for the studied samples under ambient pressure and at an applied hydrostatic pressure of about 10 kbar.

The temperature dependence of the magnetic moment \( m \) measured under an applied hydrostatic pressure of 10.4 kbar, for selected magnetic fields in the geometry \( H \parallel ab\)-plane for sample I, is presented in figure 3(a). From the above data we have determined \( T_{c2}(H = \text{const}) \), at the point where \( m(T) \) deviates from linear temperature dependence, approximating well the magnetic susceptibility in the normal state. The \( T_{c2}(H) \) data determined in this manner for various fields allowed us to plot \( H_{c2}(T) \) dependences for \( H \parallel c\)-axis and \( H \parallel ab\)-plane for the studied samples under ambient pressure and at a hydrostatic pressure of about 10 kbar.

The temperature dependences of the upper critical field for \( H \parallel c\)-axis (\( H_{c2}^{c} \)) and \( H \parallel ab\)-plane (\( H_{c2}^{ab} \)) for sample I at ambient pressure and under a hydrostatic pressure of 10.4 kbar are shown in the upper panel of figure 3(b). A significant increase of the upper critical field under pressure is clearly visible in this figure. Mainly, it results from the increase of \( T_c \) by about 7 K under a pressure of 10.4 kbar. However, a significant increase of the slope \(-dH_{c2}/dT\) in the linear part of the \( H_{c2}(T) \) dependence is observed for higher fields. For lower fields, in the vicinity of \( T_c \), one can notice strong curvature. For \( H \parallel c\)-axis, in the field range between 10 and 50 kOe, we have \(-dH_{c2}/dT = 15(1)\) kOe K\(^{-1}\) at ambient pressure, which rises up to 22(3) kOe K\(^{-1}\) under 10.4 kbar. In the case of \( H \parallel ab\)-plane, an increase from 22(2) kOe K\(^{-1}\) \((P = 0\) kbar) up to 34(3) kOe K\(^{-1}\) under a pressure of 10.4 kbar is observed. The anisotropy of the slope \(-dH_{c2}/dT\) in moderate fields, being equal to about 1.5 for sample I under ambient pressure and under a pressure of 10.4 kbar, correlates quite well with the anisotropy of the penetration depth in the vicinity of \( T_c \) for a single crystal of FeTe\(_{0.55}\)Se\(_{0.5}\) investigated by Bendele \textit{et al} [29] under ambient pressure. The estimation of the zero-temperature value \( H_{c2}(0) \) by extrapolation of the present data, covering a limited temperature range, down to low temperatures [31] is not obvious because of the strong curvature of \( H_{c2}(T) \) and the possibly multiband nature of the superconductivity. Nevertheless, assuming that the value of \( H_{c2}(0) \) is proportional to \( T_c \) and to \(-dH_{c2}/dT\), determined in a relatively wide field range above the strong curvature of \( H_{c2}(T) \) in the vicinity of \( T_c \) [31], we can estimate a change of \( H_{c2}^{c}(0) \) from 150 kOe under ambient pressure to 325 kOe under a hydrostatic pressure of 10.4 kbar, which corresponds to a decrease of the zero-temperature coherence length \( \xi_{ab} \).
The temperature dependence of the magnetic moment measured for sample I under an applied hydrostatic pressure of 10.4 kbar, shown for selected magnetic fields for $H \parallel ab$-plane. (b) The temperature dependence of the upper critical field for sample I for $H \parallel c$-axis and $H \parallel ab$-plane at ambient pressure and under a hydrostatic pressure of 10.4 kbar (upper panel) and for sample II for $H \parallel c$-axis and $H \parallel ab$-plane at ambient pressure and under a hydrostatic pressure of 11.3 kbar for $H \parallel c$-axis and 9.3 kbar for $H \parallel ab$-plane (lower panel).

Figure 3. (a) The temperature dependence of the magnetic moment from about 4.7 to 3.2 nm, according to the relation [32]

$$H_{c2}^c = \frac{\Phi_0}{2\pi \xi_{c2}^2},$$

where $\Phi_0$ is the elementary flux quantum and $\xi_{c2}$ is the coherence length in the $ab$-plane.

The lower panel of figure 3(b) presents the temperature dependences of the upper critical field for $H \parallel c$-axis and $H \parallel ab$-plane for sample II at ambient pressure and under a hydrostatic pressure of about 10 kbar. The strong curvature of $H_{c2}(T)$ in the vicinity of $T_c$ noticed for sample I is much more suppressed for the sample with the sharper transition to the superconducting state (sample II). The higher values of $H_{c2}$ observed for $H \parallel c$-axis under an applied pressure of 11.3 kbar for this sample than those recorded for $H \parallel ab$-plane under a pressure of 9.3 kbar are due to the difference in the applied pressure and, therefore, to the difference in $T_c$ values. The slope $-dH_{c2}/dT$, determined in the field range between 10 and 50 kOe, is much larger for the sample with a sharp transition to the superconducting state (sample II). For sample II, we found values of $-dH_{c2}/dT$ equal to about 45(5) kOe K$^{-1}$ for $H \parallel c$-axis and to about 50(5) kOe K$^{-1}$ for $H \parallel ab$-plane, indicating a much smaller value of the upper critical field anisotropy. Furthermore, the slope $-dH_{c2}/dT$ is within experimental accuracy unchanged under pressure, suggesting that the increase of $H_{c2}$ under pressure is directly related to the changes in $T_c$ under pressure only. The presented data lead to an estimation of a change of $H_{c2}(0)$ from 450 kOe under ambient pressure to 690 kOe under a hydrostatic pressure of 11.3 kbar, which corresponds to a decrease of zero-temperature $\xi_{c2}$ from about 2.7 to 2.2 nm. The relatively large values of $H_{c2}$ and its small anisotropy for sample II most likely result from the extended amount of defects in the structure evidenced by wide x-ray peaks [23] and, therefore, they may not correspond to intrinsic $H_{c2}$ values. On the other hand, the sample with a larger amount of defects is characterized by stronger interband scattering and the appearance of sufficiently strong interband scattering is an essential for enhanced superconducting state properties.

The temperature dependence of the lower critical field $H_{c1}$ was studied by following the field $H_{c1}^s$ for which the first vortices start to penetrate the sample at its surface, that is directly related to $H_{c1}$ [29]. The field $H_{c1}$ was estimated according to the procedure introduced in [33] and discussed in [29]. The quantity $(BV)^{1/2}$ was calculated from the measured magnetic moment $m = MV$ and plotted as a function of the internal magnetic field $H_{int} = H_{ext} - DM$, where $H_{ext}$ denotes the external magnetic field (see the inset to the upper panel of figure 4(a)). Here, $B$ denotes the magnetic induction and $V$ is the sample volume. Since $B = 4\pi M + H_{int} = 4\pi m/V + H_{int} = 0$ in the Meissner state, it is possible to determine, from the data of $(m/H_{int})$, the field $H_{c1}^s$ above which this equality is invalid. Hence, the magnetic induction $B$ empirically scales as the square of $H$ above $H_{c1}^s$, and a plot of $(BV)^{1/2}$ as a function of $H_{int}$ allows a straightforward determination of $H_{c1}^s$. The sudden increase from zero occurs due to the penetration of vortices at $H_{c1}^s$. For the case of weak bulk pinning, the surface barrier may play a crucial role and determine the first field of flux penetration and the irreversibility line [34–36]. The impact of the surface barrier leads to asymmetric $M(H)$ loops.
that entry and exit of magnetic flux and, therefore, we assume the descending branch is in such a case almost horizontal. For presented in semilogarithmic scale for sample II. ∥\text{H}\parallel c\text{-axis and }\text{H}\parallel ab\text{-plane determined at ambient pressure and under a hydrostatic pressure of 10.4 kbar (upper panel) and for sample II at ambient pressure and under a hydrostatic pressure of 11.3 kbar for }\text{H}\parallel c\text{-axis and 9.3 kbar for }\text{H}\parallel ab\text{-plane. (b) The reduced temperature dependences of the lower critical field for both field configurations correlate very well with the data presented in figure 4(b) in semilogarithmic scale. The anisotropy of the lower critical field (γHc1) does not increase under applied hydrostatic pressure; the inset to figure 4(b) rather indicate a slight decrease of γHc1. In order to extract the values of the magnetic penetration depth from the measured values of Hc1, the following basic relations were applied [32]:

\[ H_{c1}^* = \frac{\Phi_0}{4\pi\lambda_{ab}^2} \ln(\kappa^*_{ab}) + 0.5, \]

\[ H_{c1}^{ab} = \frac{\Phi_0}{4\pi\lambda_{c}^2} \ln(\kappa^{ab}) + 0.5. \]

Here, \(\lambda_{ab}\) and \(\lambda_{c}\) denote the magnetic penetration depths related to the superconducting current flowing in the \(ab\)-plane and along the \(c\)-axis, respectively, \(\xi_{ab}\) and \(\xi_{c}\) are the corresponding coherence lengths, and \(\kappa^*_{ab} = \lambda_{ab}/\xi_{ab}\) and \(\kappa^{ab} = (\lambda_{ab}\lambda_{c}/\xi_{ab}\xi_{c})^{1/2}\) are the corresponding Ginzburg–Landau parameters. The zero-temperature values of \(\xi_{ab}(0)\) and \(\xi_{c}(0)\) at ambient pressure and under hydrostatic pressure were derived from the values of \(H_{c1}^*\) and \(H_{c1}^{ab}\) extrapolated to zero temperature for both field configurations. Then, for sample II, the following zero-temperature values of the magnetic penetration depths at ambient pressure were obtained: \(\lambda_{ab}(0) \approx 400(50)\) nm and \(\lambda_{c}(0) \approx 900(200)\) nm. These values are in a very good agreement with the values determined by \(\mu\)SR measurements [29]. The corresponding zero-temperature values of the magnetic penetration depth at a hydrostatic pressure of about 10 kbar are as follows: \(\lambda_{ab}(0) \approx 180(20)\) nm and \(\lambda_{c}(0) \approx 320(50)\) nm. Obviously, the state is characterized by larger values of Hc1 (sample II). This means that the penetration depth for this sample is smaller and the superconducting carrier density is larger than that of the high crystallographic quality sample (sample I).

The obtained data additionally indicate that the structural disorder originating from the kinetics of the crystal growth process influences the superconducting properties. In particular, our data support the observation that the ions’ inhomogeneous spatial distribution enhances the superconductivity. Since the observed improvement of superconducting state properties is correlated with the suppression of the curvature of \(H_{c2}(T)\) in the vicinity of \(T_c\), one may suppose that an increase of interband scattering is directly responsible for the improvement of the superconducting properties in the studied multiband superconductor.

From the data presented in the lower panel of figure 4(a), the extrapolated zero-temperature values for sample II were found to be \(H_{c1}^{ab}(0) = 27(5)\) Oe and \(H_{c1}^{c}(0) = 56(8)\) Oe at ambient pressure and \(H_{c1}^{ab}(0) = 250(30)\) Oe under a pressure of 11.3 kbar while \(H_{c1}^{ab}(0) = 150(30)\) Oe under a pressure of 9.3 kbar. The zero-temperature values of the lower critical field for both field configurations correlate very well with the values obtained by Bendele et al for single crystals of identical nominal compositions [29]. The Hc1 increases significantly under applied external pressure for all studied temperatures. The reduced temperature dependences of the lower critical field at ambient pressure and under a hydrostatic pressure of 11.3 kbar for \(H\parallel c\)-axis and 9.3 kbar for \(H\parallel ab\)-plane are presented in figure 4(b) in semilogarithmic scale. The \(\lambda_{ab}\) and \(\lambda_{c}\) denote the magnetic penetration depths related to the superconducting current flowing in the \(ab\)-plane and along the \(c\)-axis, respectively, \(\xi_{ab}\) and \(\xi_{c}\) are the corresponding coherence lengths, and \(\kappa^{ab} = (\lambda_{ab}\lambda_{c}/\xi_{ab}\xi_{c})^{1/2}\) are the corresponding Ginzburg–Landau parameters. The zero-temperature values of \(\xi_{ab}(0)\) and \(\xi_{c}(0)\) at ambient pressure and under hydrostatic pressure were derived from the values of \(H_{c1}^*\) and \(H_{c1}^{ab}\) extrapolated to zero temperature for both field configurations. Then, for sample II, the following zero-temperature values of the magnetic penetration depths at ambient pressure were obtained: \(\lambda_{ab}(0) \approx 400(50)\) nm and \(\lambda_{c}(0) \approx 900(200)\) nm. These values are in a very good agreement with the values determined by \(\mu\)SR measurements [29]. The corresponding zero-temperature values of the magnetic penetration depth at a hydrostatic pressure of about 10 kbar are as follows: \(\lambda_{ab}(0) \approx 180(20)\) nm and \(\lambda_{c}(0) \approx 320(50)\) nm. Obviously, the

![Figure 4.](image-url)
estimated low-temperature anisotropy of the penetration depth for FeTe$_{0.5}$Se$_{0.5}$ under hydrostatic pressure is significantly smaller than that under ambient pressure. Furthermore, the obtained data suggest that the anisotropy of $\lambda$ does not increase with decreasing temperature, which is typical for chalcogenides at ambient pressure. However, the obtained data are insufficient to make conclusive statements concerning the temperature dependence of the anisotropy of the penetration depth in FeTe$_{0.5}$Se$_{0.5}$ under pressure. A summary of the changes of the thermodynamic parameters under pressure for both studied samples is given in table 1.

Uemura et al [37] have found an empirical relation between the zero-temperature superconducting carrier density $\rho_s(0)$ and $T_c$ which seems to be generic for various families of cuprate high-temperature superconductors (Uemura plot). This ‘universal’ relation $T_c(\rho_s)$ has the following features: with increasing carrier doping $T_c$ initially increases linearly ($T_c \propto \rho_s(0)$), then saturates, and finally is suppressed for high carrier doping. It is interesting to check how the Uemura relation holds for iron-based superconductors subjected to hydrostatic pressure. For this reason, $T_c$ versus $\lambda_{ab}^{-2}(0)$ is plotted in figure 5 for a selection of various Fe-based superconductors investigated so far [29, 38–48], together with the pressure impact on the positions of both FeTe$_{0.5}$Se$_{0.5}$ samples investigated in this work. The figure was prepared using the values of $T_c(\lambda_{ab}^{-2}(0))$ obtained for sample I at ambient pressure and under a hydrostatic pressure of 10.4 kbar and for sample II at ambient pressure and under a hydrostatic pressure of 11.3 kbar. The Uemura relation observed for underdoped cuprates is included for comparison as a dashed line for hole doping and as a dotted line for electron doping. The penetration depth values obtained under ambient pressure locate the studied samples in the area of hole-doped compounds. An application of hydrostatic pressure of about 10 kbar shifts the positions of studied samples in the diagram of $T_c(\lambda_{ab}(0))$ towards the area of electron-doped compounds, instead of a shift along the line denoting hole-doped compounds. The effect is very well visible for sample II placed almost ideally on the line denoting hole-doped compounds at ambient pressure as well as on the line denoting electron-doped compounds under a hydrostatic pressure of 11.3 kbar. Sample I, despite its essentially identical value of $T_c$ to sample II, is characterized by a much higher value of $\lambda_{ab}(0)$ both at ambient and under hydrostatic pressure, and therefore its position in the Uemura plot is shifted towards the lower $\lambda_{ab}^{-2}$ values as compared to those expected for hole-doped and electron-doped compounds, respectively. Obviously, for both studied samples the external pressure affects the density of superconducting carriers. However, it may cause also an induction of magnetic phase, similar to that reported by Bendele et al [49] in FeSe crystal, manifested by the $T_c(P)$ dependence not going along the

| Quantity | Sample I | 0 kbar | 10.4 kbar | 0 kbar | 11.3(∗) or 9.3(∗∗) kbar |
|----------|----------|--------|-----------|--------|------------------------|
| $T_c$ (K) | 14.2(2)  | 21.2(2) | 14.2(2)  | 22.0(2)  |                        |
| $-dH^c_{T_0}/dT$ (kOe K$^{-1}$) | 15(1)    | 22(3)  | 45(5)    | 45(5)   |                        |
| $-dH^c_{T_1}/dT$ (kOe K$^{-1}$) | 22(2)    | 34(3)  | 50(5)    | 50(5)   |                        |
| $H^c_{T_0}$ (kOe) | 150(10)  | 325(45)| 450(50)  | 690(80) |                        |
| $H^c_{T_1}$ (kOe) | 220(20)  | 505(45)| 500(50)  | 770(80) |                        |
| $H^c_{T_1}$ (eV) | 22(2)    | 34(3)  | 50(5)    | 50(5)   |                        |
| $\xi_{ab}(0)$ (nm) | 4.7(2)   | 3.2(2) | 2.70(15) | 2.20(15)|                        |
| $\lambda_{ab}(0)$ (nm) | 740(80)  | 275(30)| 400(50)  | 180(20) |                        |
| $\xi_{c}(0)$ (nm) | 3.9(2)   | 2.55(15)| 2.55(15)| 2.05(15)|                        |
| $\lambda_{c}(0)$ (nm) | 850(180)| 380(70)| 900(200)| 320(50)|                        |
| $\kappa_{c}$ (°C) | 160(20)  | 85(15) | 150(20)  | 80(15)  |                        |
| $\kappa_{ab}$ (°C) | 185(40)  | 115(30)| 230(50)  | 115(30) |                        |

Table 1. The pressure impact on the thermodynamic parameters describing the superconducting state for both investigated single crystals of FeTe$_{0.5}$Se$_{0.5}$.

Figure 5. The pressure impact on the position of FeTe$_{0.5}$Se$_{0.5}$ in the Uemura plot of a selection of Fe-based high-temperature superconductors (after [29]). The arrows indicate changes of the positions in the plot of the investigated crystals of significantly different crystallographic qualities when subjected to hydrostatic pressures of 10.4 kbar (sample I) and 11.3 kbar (sample II). The Uemura relation observed for underdoped cuprates is included for comparison as a dashed line for hole doping and as a dotted line for electron doping.
hole-doped compounds line. Importantly, we noticed a more pronounced increase in the superconducting carrier density under pressure than that in the critical temperature, which may indicate the appearance of a mechanism limiting the increase of $T_c$ with pressure. However, we should note that the change of lattice constants under pressure leads to a change of the superconducting carrier effective mass which affects the values of $\lambda_{ab}(0)$.

### 3.3. The critical current density

The hysteresis loops of the studied single crystals were recorded at various temperatures in a magnetic field applied along both $H \parallel c$-axis and $H \parallel ab$-plane at ambient pressure and under a hydrostatic pressure of about 10 kbar. Figure 6 presents typical data recorded for sample I for $H \parallel c$-axis at 5 K at ambient pressure and at 7.3 K under a hydrostatic pressure of 10.4 kbar, i.e., at the same reduced temperature of 0.35$T_c$. Using Bean’s model [50, 51], for the sample of rectangular shape, one can estimate the superconducting critical current density according to the formula

$$j_c(H) = \frac{20\Delta M(H)}{a(1 - \frac{H}{H_c^2})}. \quad (4)$$

Here, $\Delta M$ (in Gauss) is the width of the hysteresis loop (see figure 6), $a$ and $b$ are the sample dimensions (in cm) in the plane perpendicular to the applied magnetic field and the critical current density is in A cm$^{-2}$. The magnetic field dependence of the critical current density for sample I at ambient pressure and under a hydrostatic pressure of 10.4 kbar, calculated according to equation (4), for all of the studied temperatures in the magnetic field geometries $H \parallel c$-axis and $H \parallel ab$-plane is presented in figure 7(a) (upper and middle panels). We note the relatively small value of the estimated critical current density, $j_c$, as compared to those observed in single-crystalline iron pnictides [52]. However, the obtained $j_c$ values are not surprising since it has been shown that FeTe$_{0.5}$Se$_{0.5}$ may exhibit the coexistence of two tetragonal phases [9, 26, 27]. The presence of such phases lowers the transport current density as phase separation boundaries prevent to develop a global circulating current [29]. This leads to a relatively low value of the magnetic critical current density when calculated taking into account the diameter of the sample. Furthermore, both the upper and the lower critical fields for sample I are quite small in comparison to those for sample II (see figures 3(b) and 4(a) and table 1) and the pinning is expected to be proportional to the thermodynamic critical field. The field dependence of the increase of the critical current density under pressure, i.e., of the ratio of critical current densities under a hydrostatic pressure of 10.4 kbar and at ambient pressure at reduced temperatures of 0.35, 0.55, and 0.69$T_c$ in magnetic fields $H \parallel c$-axis and $H \parallel ab$-plane is presented in the lower panel of figure 7(a). The critical current density strongly increases under pressure by at least one order of magnitude, for $H \parallel c$-axis and $H \parallel ab$-plane, for all investigated reduced temperatures and in the full magnetic field range (lower panel of figure 7(a)). It can be explained by an improvement of the effectiveness of small defects in the sample subjected to pressure, because of a decrease of the coherence length under pressure, and by an increase of the thermodynamic critical field under pressure due to the increase of both the lower and the upper critical fields. The influence of pressure on $j_c$ is evidently stronger at higher magnetic fields, by up to two orders of magnitude (lower panel of figure 7(a)). This is not surprising since a significant increase of $H_{c2}$ under pressure was noted too.

The magnetic field dependence of the critical current density for sample II at ambient pressure and under a hydrostatic pressure of 11.3 kbar in the magnetic field geometries $H \parallel c$-axis and 9.3 kbar in the $H \parallel ab$-plane geometry for all of the studied temperatures is presented in figure 7(b) (upper and middle panels). The field dependence of the increase of the critical current density under pressure is presented in the lower panel of figure 7(b). Sample II is characterized by a significantly enhanced critical current density at ambient pressure, as compared to sample I, because of the extended amount of defects in the structure, evidenced by relatively wide x-ray peaks. Consequently, the increase of the critical current density under pressure is strongly reduced for sample II, especially in the geometry $H \parallel c$-axis and at low temperatures, where the initial critical current density is the highest.

### 4. Conclusions

Magnetic studies at ambient pressure and under hydrostatic pressure were performed for single crystals of FeTe$_{0.5}$Se$_{0.5}$ in order to investigate the impact of pressure on the basic parameters of the superconducting state. We compared the influence of hydrostatic pressure on the properties of two crystals with significantly different amounts of defects. We found pressure-induced enhancement of all the investigated parameters. Furthermore, we noted that the application of hydrostatic pressure does not increase the anisotropy of
the superconducting state parameters. However, a more pronounced increase in the superconducting carrier density under pressure than that in the critical temperature was found, indicating the appearance of a mechanism limiting the increase of $T_c$ with pressure.

Comparison of the pressure impact on the superconducting properties of two samples with different amounts of defects leads to the following conclusion: the significant suppression of the strong curvature of $H_{c2}(T)$ in the vicinity of $T_c$ for the sample with an extended amount of defects indicates the increasing interband scattering as a result of the increasing structural inhomogeneity. Since the suppression of the curvature of $H_{c2}(T)$ in the vicinity of $T_c$ is correlated with an observed improvement of the superconducting state properties one may suppose that an increase of interband scattering is directly responsible for the improvement of the superconducting properties in the studied multiband superconductor. It may explain the origin of the relatively poor superconducting state properties of the single crystals of better crystallographic quality.

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