Doubling of the critical temperature of FeSe observed in point contacts

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Rise in superconducting critical temperature T_c more than two times (exceeding $20\,K$) is discovered in point-contacts created between iron-chalcogenide FeSe single crystal and Cu. The possible reasons of such T_c increase in point-contacts are discussed. The most probable cause for this may be the interfacial carriers doping and/or interfacial enhanced electron-phonon interaction.

I. INTRODUCTION

The superconducting (SC) compound FeSe, having the simplest crystal structure among other SC iron chalcogenides and pnictides, has attracted a great interest during the last years. Primarily, this is connected with the possibility of a large alteration of their SC critical temperature T_c. Thus, the moderate T_c of about $8\,K$ in bulk FeSe[1] increases drastically up to 27 K under pressure[2] and achieves even a maximum of $36.7\,K$ at $8.9\,GPa$[3]. Such T_c enlargement is unlikely to be found for any other SC material. On the other hand, T_c climbs up to tremendous $109\,K$ in the case of a FeSe monolayer on $SrTiO_3$, as it has been shown by means of in situ electrical transport measurements[4]. After that, various methods to increase T_c were utilized. One of them is doping of the FeSe topmost layer by excess electrons by covering the surface using alkali metals[5] or applying the liquid-gating technique[6]. However, as it was shown recently[7] such doping has only a moderate effect, increasing T_c up to 20 K of FeSe bulk crystal. That is, substrate-interfacial effects play the main role in increasing T_c in the case of FeSe monolayer on $SrTiO_3$, likely due to interface enhanced electron-phonon interaction. Although FeSe monolayer has very high T_c, they survive, so far, only in-situ at high vacuum condition, while using of a protection layer suppresses high-T_c superconductivity in FeSe monolayer[4]. Recently, an increase of T_c almost twice than that of the bulk crystals, was reported for atmosphere-stable FeSe films with a practical thickness of about several hundred nanometers[8]. This increase has been explained by proper tuning the Fe-vacancy disorders via changing the Fe/Se ratio. In this communication, we report about the observation of more than doubling of T_c onset in point-contacts (PCs) created on a bulk FeSe single crystal. We believe that our results provide helpful information in order to understand in more detail the role of the interface to modify the properties of superconducting FeSe.

II. EXPERIMENTS AND RESULTS

The plate-like single crystals (flakes) of $FeSe_{1-x}$ ($x=0.04\pm0.02$, #CD-946) superconductor were grown in evacuated quartz ampoules using $AlCl_3/KCl$ flux technique in a permanent temperature gradient as described in Ref.[9]. Resistivity and magnetization measurements revealed a SC transition temperature T_c up to $9.4\,K$[9] and an onset of superconductivity at about $10\,K$.

PCs were established by touching of a sharpened thin Cu wire ($\varnothing=0.2\,mm$) to the ab-plane of FeSe cleaved by a scalpel at room temperature or contacting by the wire an edge of the plate-like samples. Thus, we have measured heterocontacts between normal metal Cu and the FeSe crystal mostly along two directions.

The differential resistance $dV/dI(V) = R(V)$ of PC was recorded by sweeping the dc current I on which a small ac current i was superimposed using a standard lock-in technique. The measurements were performed in the temperature range from $3\,K$ up to $25\,K$. No principal difference in $dV/dI(V)$ data was observed for "plane" or "edge" PC geometry, because $dV/dI(V)$ differ more significantly from one PC to another.

Typical $dV/dI(V)$ data for PCs with different resistance are shown in Fig. 1 in our previous publication[10]. For low-Ohmic PCs with resistance up to several Ohms the main feature in $dV/dI(V)$ is a pronounced sharp zero-bias minimum[10]. For the overwhelming majority of PCs, independently, either $dV/dI(V)$ demonstrate additionally occasional Andreev-reflection like features or not[14] (see, e.g. Figs. 4 & 5 in Ref.[10]), this minimum and accompanying side maxima disappear at temperature around $10\,K$[15]. This range was a bit (1–2 K) higher for some of the PCs. Unexpectedly, we have found PCs (one PC created by "edge" geometry and one PC created on ab-plane), where SC features were observed to break down only above $20\,K$, as it is shown in Figs. 1 & 2. The statistic of the variation of T_c^{onset} with the PC resistance is shown in Fig. 3.

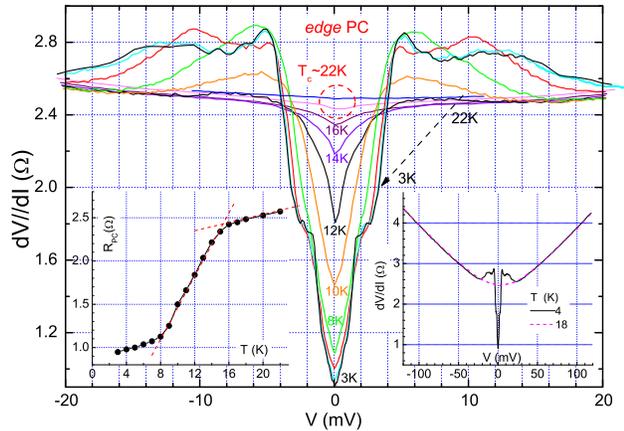Thereby, we claim to observe a doubling of the local

FIG. 1: (Color online) Evolution of dV/dI curves with temperature for a PC created by touching of an edge of a FeSe crystal by a Cu wire. The upper two curves marked by the circle are measured at 18 and 22 K (top curve). Right inset shows two dV/dI curves measured at 4 and 18 K for a larger bias. Left inset shows the dependence of the PC resistance at zero-bias versus temperature. Dotted lines are given to guide the eye.

FIG. 2: (Color online) Evolution of dV/dI curves with temperature for PC created by touching of ab-plane of FeSe flake by a Cu wire. The upper two curves marked by circle are measured at 20 and 22 K (top curve). Right inset shows dV/dI measured at 3, 12 and 22 K for a larger bias. Left inset shows dependence of the PC resistance at zero-bias versus temperature. Full (red) circles show corrected PC resistance after subtracting from resistance increase above 12 K (as it is seen from the shifting of dV/dI "background" at larger bias) coming, likely, from the bulk. Dotted line is given to guide the eye.

T_c^{onset} in a restricted geometry of the FeSe crystal, which is determined by the PC size. The latter was estimated in our previous paper[10] for a PC with similar resistance to be in the range of 0.1-1μm. The large uncertainty in the determination of the PC size is due to the unknown residual resistivity of FeSe at the interface with Cu and due to the heterocontact geometry, where the partial filling of the PC volume by one (FeSe) or another (Cu) material is unknown.

III. DISCUSSION

The first thought that comes to mind about increasing of T_c in PC is that it is connected with a pressure effect. Indeed, the small size of a PC (or more precisely speaking, the small contact area, which, in general, can be larger than that of metallic contact itself due to, e.g., surface oxides), which can be in the order of a few microns, make it possible to cause large pressure (by mechanical creation of PC) within the PC core. According to Fig. 4 in Ref.[2], to reach the onset critical temperature of about 20 K, the pressure should exceed 1 GPa. At the same time, we believe that metallic Cu wire cannot produce a pressure larger than the yield strength of Cu. The latter reaches only about 0.07 GPa[16] and cannot be much larger at low temperature.

Another observation that contradicts the pressure explanation of the T_c increase comes from Fig. 3. Intuitively it is clear that the pressure in a PC is expected to be larger for PCs with larger resistance or respectively
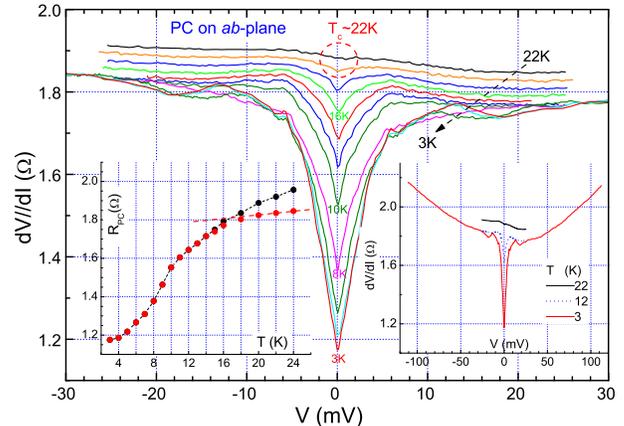
smaller size. Contrary, as shown in Fig. 3, two PCs with smaller resistance (larger size) exhibit a two-times higher T_c^{onset} than the other PCs. Probably, the pressure effect is responsible mainly for the T_c^{onset} scattering between 10 and 14 K as seen in Fig. 3.

An enhancement of T_c in PCs was observed also in Co-doped Ba-122[12] and in FeTe$_{0.55}$Se$_{0.45}$[13] iron-based superconductors. In both cases this enhancement was about 30%, that is much less than we observe. In the first case, the authors suppose the formation of phase-incoherent quasiparticle pairs at a temperature well above T_c arising from strong fluctuation of the phase of the complex superconducting order parameter. In the second case, the authors assume also novel quasiparticle scattering due to strong antiferromagnetic spin fluctuations. As to our case, we do not believe that such kind of fluctuations is able to increase T_c on 100%.

Different scenario to explain the T_c rise in FeSe films may be a fortunate arrangement of iron vacancies somewhere at the interface, as assumed in Ref.[8]. However, T_c^{onset} in that FeSe films was found to increase only up to about 15 K that is 5 K lower than in our case. Nevertheless, it cannot be excluded also the joint effect of pressure and iron vacancy arrangement, that might result in 20 K onset superconductivity in our PCs. However, high pressure in PC will result rather in disarrangement of the Fe vacancies.
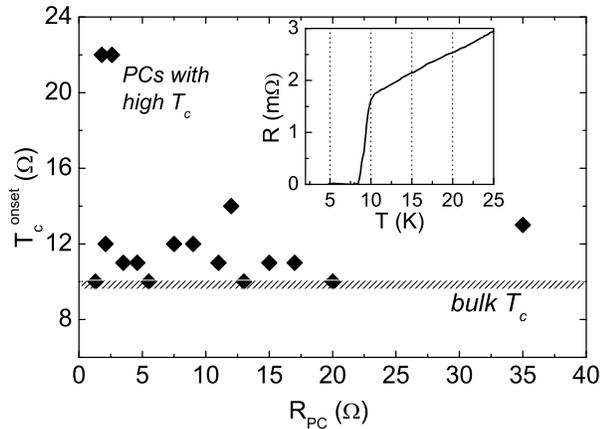
Alternatively, the observed T_c increase may be an

FIG. 3: Statistic data of T_c^{onset} variation for PCs with different resistance. Horizontal stripe marks T_c^{onset} for a bulk FeSe taken from the resistance data shown in the inset. Inset: resistance versus temperature of FeSe sample from Ref.[9].

interfacial effect due to additional doping[7] and/or interface-enhanced electron-phonon coupling[4] from the side of the normal metal. Thus, a real understanding of the observed T_c rise in PCs on the base of FeSe is a challenging task. In this case, the restricted PC geometry and interfacial effects can play a decisive role.

IV. CONCLUSION

In summary, we have investigated the nonlinear conductivity of PCs on the base of FeSe single crystals due to the transition into the SC state. We found that SC features in the differential resistance $dV/dI(V)$ persist up to 20 K for some PCs. Such doubling of the local critical temperature in FeSe cannot be explained only by pressure effects in the PC. As a possible explanation a suitable arrangement of iron vacancies is discussed along with the presence of pressure. Apparently, the underlying physical nature of the observed effect can be understood by taking into account the restricted geometry of the PC core and interfacial effects.

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[14] The presence or absence of the characteristic Andreev-reflection features in dV/dI curves allows to conclude about regime (ballistic, diffusive or thermal)[11] of current flow in superconducting PCs. In general, the critical temperature has no relation to this regime of current flow through a PC. Moreover, the heating and critical current effects do not play a role at T_c^{onset} determination, since the disappearance of superconductivity in PC we can monitor watching the minimum in dV/dI at zero bias (or at zero current). On the other hand, the critical temperature itself can be a criteria of the PC quality. When it is lower than the bulk value, it means that the PC quality is low.

[15] As a critical temperature onset T_c^{onset} in PC, we took the temperature at which the zero-bias minimum disappears, as was marked by the dotted circle in Figs. 1&2.

[16] See "The Engineering Tool Box" web site http://www.engineeringtoolbox.com/young-modulus-d_417.html