Superconducting gaps in FeSe studied by soft point-contact Andreev reflection spectroscopy

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FeSe single crystals have been studied by soft point-contact Andreev reflection spectroscopy. Superconducting gap features in the differential resistance $dV/dI(V)$ of point contacts such as a characteristic Andreev reflection double-minimum structure have been measured versus temperature and magnetic field. Analyzing dV/dI within the extended two-gap Blonder-Tinkham-Klapwijk model allows one to extract both the temperature and magnetic field dependence of the superconducting gaps. The temperature dependence of both gaps is close to the standard BCS behavior. Remarkably, the magnitude of the double-minimum structure gradually vanishes in magnetic field, while the minima position only slightly shifts with field, indicating a weak decrease of the superconducting gaps. Analyzing the $dV/dI(V)$ spectra for 25 point contacts results in the averaged gap values $\langle\Delta_L\rangle = 1.8 \pm 0.4$ meV and $\langle\Delta_S\rangle = 1.0 \pm 0.2$ meV and reduced values $2\langle\Delta_L\rangle/k_BT_c = 4.2 \pm 0.9$ and $2\langle\Delta_S\rangle/k_BT_c = 2.3 \pm 0.5$ for the large (L) and small (S) gap, respectively. Additionally, the small gap contribution was found to be within tens of percent, decreasing with both temperature and magnetic field. No signatures in the dV/dI spectra were observed, testifying to a gapless superconductivity or the presence of even smaller gaps.

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I. INTRODUCTION

The binary compound FeSe, belonging to the family of iron-based superconductors, is in the focus of intense investigations nowadays. The main advantage of this material is that superconducting (SC) FeSe is the only binary compound in this family. Additionally, FeSe shows no long-range magnetic order, which might simplify the understanding of the nature of SC pairing. Furthermore, FeSe demonstrates extraordinary sensitivity of the SC properties to external pressure, chemical doping on the Fe or Se site, and to the intercalation by alkali metals (see Ref. [1] for a recent review). Besides, the critical temperature of FeSe can be enhanced by an order of magnitude by diminishing its dimensionality to a two-dimensional (2D) type monolayer. Further, an immensely small Fermi energy, which is comparable to the SC gap(s) Δ, locates the SC state of FeSe in the vicinity of the extraordinary BCS-BEC crossover [2]. Therefore, investigations of SC gap(s) in FeSe are of high interest.

Angle-resolved photoemission spectroscopy (ARPES) is the most powerful method to study the directional and band dependence of the SC gap(s). However, the resolution of ARPES measurements, which is nowadays slightly below 1 meV, does not provide sufficient accuracy for the detection of a SC gap value for superconductors with a critical temperature of about 10 K and below, as in the case of bulk FeSe. Two other spectroscopic methods, such as scanning tunneling spectroscopy (STS) [3] and point-contact Andreev reflection (PCAR) spectroscopy [4,5], have significantly better energy resolution, though both methods suffer from directional selectivity and especially from the ability to resolve electron bands.

In one of the first STS measurements on FeSe, Song *et al.* [6] reported the presence of one gap with $\Delta \sim 2.2$ meV taken as half of the peak-to-peak energy separation in tunnel dI/dV spectra. Further, Kasahara *et al.* [2] demonstrated tunnel dI/dV spectra of FeSe showing a V-shaped zero-bias minimum with the side maxima at ± 2.5 meV and shoulders at ± 3.5 meV. These features were taken as evidence of two SC gaps. Later, Watashige *et al.* [7] reported STS data and dI/dV spectra with peaks at ± 2.5 meV and shoulders outside of the main peaks at ± 3.5 meV. Moore *et al.* [8] obtained a V-shaped STS spectrum for FeSe in the low-energy range with clear peaks at $\Delta = \pm 2.3$ meV. Very recently, Jiao *et al.* [9] used the $(s + es)$ model to fit their STS data with a small s-wave gap of $\Delta_s(0) = 0.25$ meV and a large anisotropic extended s-wave gap $\Delta_{es} = \Delta_0(1 + \alpha \cos 4\Theta)$ with $\Delta_0 = 1.67$ meV and $\alpha = 0.34$, which results in a SC gap maximum of 2.24 meV and a minimum of 1.10 meV.

At the same time, one of the last ARPES studies of FeSe reported by Borisenko *et al.* [10] announced two gaps equal to 1.5 and 1.2 meV for the hole band in the center and for the electron band in the corner of the Brillouin zone, respectively. Here, we must note that recently Hong and Aberge [11] pointed out that the side peaks observed in STS measurements on compounds with a strong electron-electron correlation, such as iron-based superconductors and high-T_c superconductors, are formed by coherence-mediated tunneling under bias. Because of that, such peaks do not reflect directly the underlying density

of states (DOS) of the sample, and the gap measured between side peaks observed in STS is bigger than the SC gap observed by ARPES. This might be the reason for substantial differences in the mentioned gap values obtained by STS [2,6–8] and ARPES [10].

Turning to the Andreev reflection spectroscopy of SC gaps in FeSe, Ponomarev *et al.* [12] have detected two sets of subharmonic gap structures due to multiple Andreev reflection using break junctions with polycrystalline samples. This was taken as proof of two nodeless SC gaps $\Delta_L = 2.75 \pm 0.3$ meV and $\Delta_S = 0.8 \pm 0.2$ meV. At the same time, their result on the temperature dependence of the both gaps was curious. Later, they reported new values $\Delta_L = 2.4 \pm 0.2$ meV and $\Delta_S = 0.75 \pm 0.1$ meV using single crystals [13]. In our PCAR measurements with FeSe single crystals [14], we also extracted two gaps from measured dV/dI with gap values similar to those in STS experiments, though the contribution of the larger gap at 3.5 meV to the total PC conductivity was rather small, of the order of 10%.

As follows from all the above points, there is a challenge in determining the spectral data related to the value of the SC gaps more accurately. Besides it, there is lack of data in the literature for the temperature and especially magnetic field dependence of the SC gap(s) in FeSe. All these issues are targets of the current investigation of FeSe using the technique of PCAR spectroscopy.

II. EXPERIMENTAL DETAILS

The platelike single crystals of $FeSe_{1-x}$ ($x = 0.04 \pm 0.02$) were grown in evacuated quartz ampoules using a flux technique as described in Ref. [13]. The resistivity and magnetization measurements revealed a SC transition temperature up to $T_c = 9.4$ K. The so-called "soft" method was utilized to create point contacts, i.e., a tiny drop of silver paint was placed on the freshly cleaved surfaces of FeSe. The soft PCs were made on the *ab* plane cleaved with a scalpel or on the edge of a thin FeSe flake. We will refer to these two types of PCs as a "plane" or "edge" PC, respectively. The silver paint drop was connected to the electrical circuit by Cu, Ag, or Pt thin wires with a diameter of 0.1 mm or slightly less. The size of the silver paint drop was about several hundred microns, while the PC resistance between the silver paint drop and FeSe samples was usually in the range 0.5–10 Ω. Such resistance corresponds to a PC size of the order of several tens of nanometers [4] in the case of PC between ordinary metals. Therefore, it is assumed for our case that either there is a large number of nanometer-sized PCs, or the interface between the silver paint and FeSe has some barrier (e.g., oxide). In spite of the unknown microscopic picture of the real PC structure, the actual shape of the dV/dI characteristics is more important. As we will demonstrate below, dV/dI show typical Andreev reflection SC gap related features, which we call the double-minimum structure.

The differential resistance $dV/dI(V) \equiv R(V)$ of the 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 about 3 K to above T_c and in a magnetic field up to 15 T, applied both along the *ab* plane or the *c* axis.

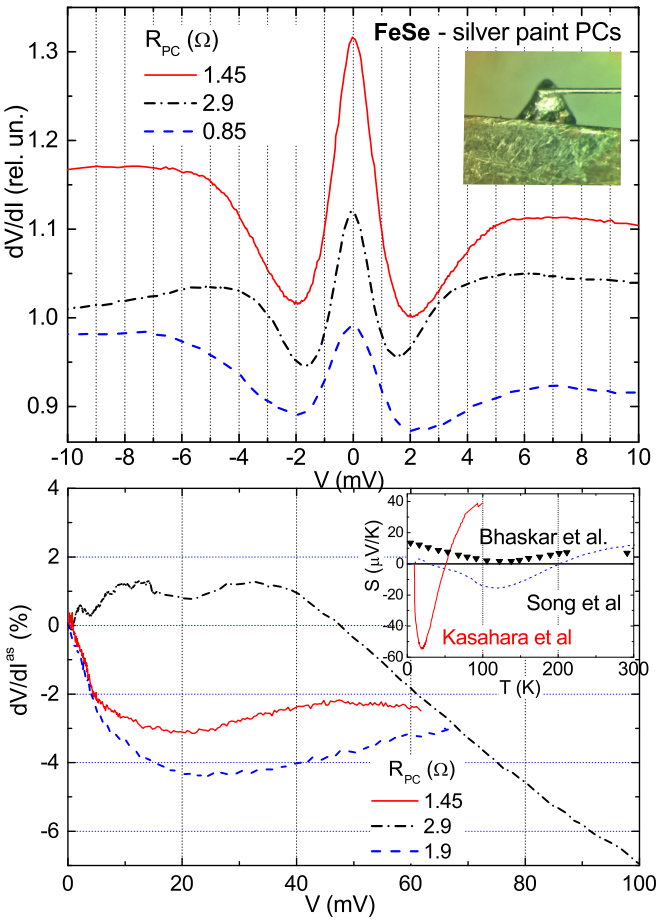

FIG. 1. Upper panel: Examples of raw dV/dI curves measured at 3 K for three "soft" PCs created by a tiny drop of silver paint on a cleaved FeSe surface. The small picture shows an image of the FeSe single crystal (triangle shape) with a drop of silver paint and a Cu wire with a diameter of 0.08 mm. Bottom panel: Antisymmetric part $dV/dI^{as}(\%) = 100[dV/dI(V > 0) - dV/dI(V < 0)]/2dV/dI(V = 0)$ of dV/dI calculated for the PCs from the upper panel. The inset shows the behavior of thermopower in single FeSe crystals according to Kasahara *et al.* [2] and FeSe polycrystals reported by Song *et al.* [17] and Bhaskar *et al.* [18].

III. RESULTS

Figure 1(a) shows the dV/dI spectra for several soft PCs, which demonstrate a characteristic double-minimum structure with the minima position between 1.5 and 2 mV, which is close to the expected SC gap value. We note the perfect reproducibility of the SC features in dV/dI, i.e., almost all of the more than 20 soft PCs with resistance in the range 0.4−5 Ω show a pronounced double-minimum structure in dV/dI. It is in contrast to our previous measurements on the same FeSe crystals using a needle-anvil geometry with tips from Cu, Ag, or W thin wires [14,15], where the double-minimum structure in dV/dI appeared very rarely. At the same time, soft PCs with a higher resistance display dV/dI with a weak zero-bias minimum or the absence of any SC features at all, similar to the needle-anvil type PCs shown in Fig. 1 in Ref. [14].

The dV/dI curves in Fig. 1(a) are asymmetric with an enhanced value at negative bias similar to our previous data

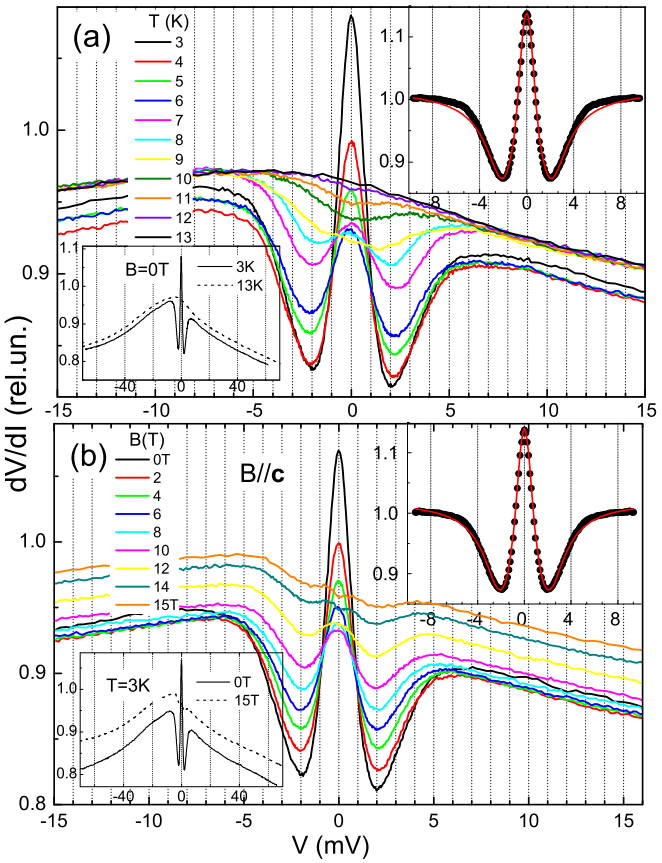

FIG. 2. (a) Temperature and (b) magnetic field evolution of the dV/dI spectra of a soft FeSe PC with a normal state resistance of 1.45 Ω. The left insets on both panels show dV/dI at a larger bias taken at low temperature and (a) above the critical temperature or (b) at maximal magnetic field. The right inset in (a) shows the fit of the symmetrized dV/dI at 3 K normalized to the normal state (see text and Table I, No. 552) by the two-gap Blonder-Tinkham-Klapwijk model, while the right inset in (b) shows the fit of the same dV/dI by the anisotropic model $\Delta = \Delta_0(1 + \alpha \cos 4\Theta)$ (see text).

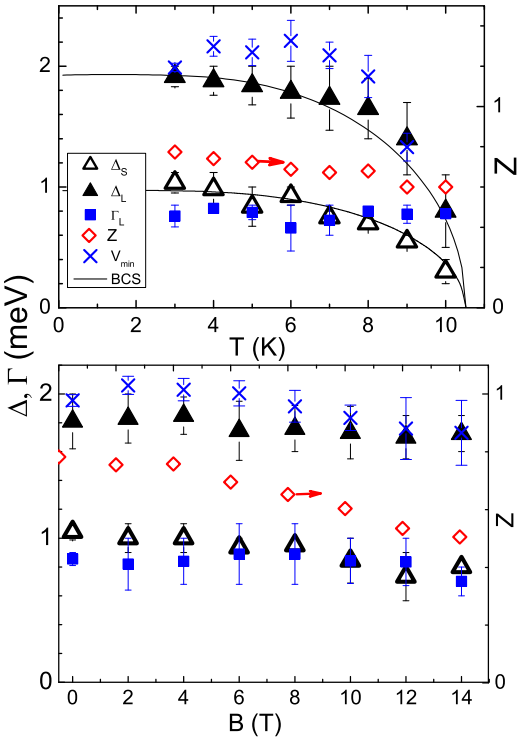

FIG. 3. Temperature and magnetic field dependencies of the fitting parameters for the PC from Fig. 2 in a two-gap approximation: Solid and open triangles are the large and small gaps Δ_L and Δ_S, respectively, Γ_L is the broadening parameter (squares), Z is the barrier parameter (diamonds), and $V_{\min}$ is the minimum position in dV/dI (crosses). Solid lines are BCS-like curves.

in Ref. [14], so that the calculated antisymmetric part of dV/dI $[dV/dI^{as}(\%) = 100[dV/dI(V > 0) - (dV/dIV < 0)]/2dV/dI(V = 0)]$ is negative [see Fig. 1(b)]. At the same time, about one third of the PCs shows positive dV/dI^{as} at low bias, as shown in Fig. 1(b).

Figure 2 demonstrates the evolution of dV/dI curves for a soft PC versus temperature and magnetic field. We used these data to determine the SC gap value and its temperature and magnetic field dependence. Therefore, we fitted [16] the dV/dI curves normalized to the normal state using the two-gap Blonder-Tinkham-Klapwijk (BTK) model (see, e.g., Refs. [4,5] for some details of the fit for different models and superconductors). An example of the fit for the dV/dI at 3 K is shown in the inset of Fig. 2(a). The fit is perfect, excluding small deviations between 4 and 8 meV, where so-called humps or side maxima occur, which arise from a non-Andreev-reflection contribution to the dV/dI spectra [19]. The results of the SC gap behavior after the fit procedure are presented in Fig. 3. The gap values at 3 K are $\Delta_L \approx 1.9$ and $\Delta_S \approx 1.0$ meV for the large (L) and small (S) gap, respectively, with an about 80% contribution to the dV/dI coming from the large gap [20]. The extracted gap values correspond to a $2\Delta/k_BT_c$ ratio of about 4.2 and 2.2 for the large and the small gap, respectively, if we use $T_c = 10.5$ K obtained from the BCS extrapolation in Fig. 3(a). The temperature behavior of both gaps is close to the BCS-like curve, while the contribution of the small gap to dV/dI spectra decreases with increasing temperature. The contribution of the small gap to dV/dI also vanishes in magnetic field, while both gap values weakly depend on the magnetic field. It is difficult to specify the critical temperature or magnetic field at which the small gap contribution disappears due to diminution and smearing of all "gap" structures with increasing temperature or magnetic field. This makes the fit procedure less unambiguous. The fit parameters for dV/dI of several PCs measured at 3 K are shown in Table I.

IV. DISCUSSION

We formed soft PCs in two ways: (a) placing a silver paint drop on the plane (flat surface) of the FeSe flake or (b) on the edge of the thin flake. In the latter case the silver paint also partially covered the flat surface, because the drop size was larger than the flake thickness. Probably due to this effect, we did not observe a big difference in the shape of dV/dI and there was also no notable difference between the gap values for the "edge" and "plane" PCs.

Here, it is appropriate to imagine the picture of how tiny PCs will be formed by dripping silver paint onto the FeSe surface.

TABLE I. Fit parameters for dV/dI of several soft PCs measured at 3 K: R_{PC} is the PC resistance, V_{min} is the minimum position in dV/dI, $\Delta_{L,S}$ is the large and small SC gaps, Γ_L is the broadening parameter, Z is the "barrier" parameter, w is the weight factor (contribution to PC conductivity) of the small gap, Δ_{ave} is the averaged gap, $\Delta_{ave} = (1-w)\Delta_L + w\Delta_S$, and S is the scaling parameter. Γ_S is taken to be zero for the small gap. Boldface type marks those PCs whose dV/dI are shown in Fig. 1.

Name	R_{PC} (Ω)	Type	V_{min}(mV)	Δ_L(meV)	Δ_S(meV)	Δ_{ave}(meV)	Z	Γ_L(meV)	w	S
No. 351	0.85	Plane	1.5	1.5	0.7	1.33	0.70	0.5	0.22	0.27
No. 373	0.67	Plane	1.75	1.73	0.9	1.51	0.77	0.55	0.26	0.19
No. 503	1.2	Plane	1.6	1.46	0.8	1.4	0.68	0.77	0.09	1.5
No. 608	3	Plane	1.7	1.53	0.8	1.49	0.68	0.87	0.06	1.18
No. 401	1.0	Edge	1.5	1.5	0.7	1.32	0.72	0.74	0.22	0.36
No. 416	2.9	Edge	1.6	1.62	0.84	1.43	0.74	0.35	0.28	0.43
No. 552	1.45	Edge	2	1.9	1.0	1.74	0.77	0.76	0.18	0.9

Let us take into consideration that, according to Ref. [21], FeSe single crystals have a huge anisotropy in resistivity between the c axis and ab plane, typically $\rho_c/\rho_{ab} \approx 500$ just above the SC transition. In such a case, we assume that the conductivity between the silver drop and the flat FeSe surface, that is along the c axis, is minor and the current flows mainly through the edges of the terraces on the surface covered by the silver paint, which opens channel(s) to the ab plane. Thus, despite the large contact area covered by the silver paint, the current flows mainly through the confined area at the edge of the terraces. As such, it does not matter whether we prepared the PC to be on the plane or edge of the FeSe flake, as in both cases the current preferably flows within the ab plane and no remarkable anisotropy is expected.

Let us discuss the details of the fit procedure. The two-gap fit uses, in general, seven parameters. Among them are two gaps $\Delta_{L,S}$, two broadening parameters $\Gamma_{L,S}$, two barriers $Z_{L,S}$, and the weight factor w. In the case when the dV/dI spectrum shows only a single double minimum, it leaves a wide scope or "too much room" for the fitting parameters and makes the fit controversial. Therefore, we shortened the number of fitting parameters by supposing equal barriers for both gaps $Z_L = Z_S$. Additionally, we supposed $\Gamma_S = 0$ by taking into account the minor small gap contribution. Thus, the number of fit parameters was reduced to five. Obviously, some variation of the extracted data is still possible even using five fitting parameters, however, in any case, the gap(s) value(s) must concentrate around the minima position of about 1.5–2 meV. This is seen also from columns No. 4–7 in Table I. The average gap values for several PCs presented in Table I are $\Delta_L = 1.6$ and $\Delta_S = 0.8$ meV, so that the large gap value is close to that measured by ARPES [10], while the small gap value is about 30% smaller than the ARPES data. On the other hand, our gap values are smaller than the gap maximum 2.24 meV and gap minimum 1.1 meV reported in Ref. [9]. However, the gap ratio Δ_L/Δ_S is close to 2 in both cases [22]. At the same time, we did not observe gap features in dV/dI similar to $\Delta_S = 0.25$ meV, as reported in Ref. [9], or gapless dV/dI behavior (such as the single V-shaped zero-bias dV/dI minimum). All of the dV/dI data from our soft PCs demonstrate a zero-bias maximum, as shown, e.g., in Fig. 1(a).

The temperature dependence of the large gap is close to the BCS behavior. At the same time the large gap is only weakly field dependent. The latter is in line with the observed minima positions in dV/dI, which are only slightly reduced in magnetic field, despite the overall suppression of the double-minimum structure. We observed a similar weak magnetic field dependence of the SC gap for another multiband superconductor from the nickel-borocarbide family, namely, $TmNi_2B_2C$ [23]. There, the possible interpretation of the observed gap behavior versus magnetic field was related to a multiband scenario. Additionally, electronic DOS modifications in the mixed state and vortex pinning near the contact interface were suggested. However, such magnetic field gap behavior is still not completely understood.

We fitted our data also by the anisotropic gap model with $\Delta = \Delta_0(1 + \alpha \cos 4\Theta)$ [9]. Additionally, we included in this model a smearing parameter $\Gamma = \Gamma_0(1 + \alpha \cos 4\Theta)$ as well. The description of the experimental data by this model is also fine [see Fig. 2(b) inset]. The fit results in $\Delta = 1.42$ meV, $\alpha = 0.6$, $Z = 0.81$, and $\Gamma = 0.53$ meV. The extracted temperature dependence of $\Delta(T)$ almost perfectly follows the BCS dependence. However, the monotonic increase of α with temperature up to the maximal value 1 close to T_c is not physically reasonable in this case, whereas on the other hand Γ_0 goes down to zero. If we try to keep α more or less constant, the fit becomes worse and Δ_0 slowly increases with temperature before a drop at approaching T_c. So, our conclusion is that the anisotropic α model is less compatible with our data.

Let us discuss the statistical data after analyzing all 25 soft PCs [22]. The position of V_{min} agglomerates in the range between 1.5 and 2 mV with an average value of $\langle V_{min}\rangle = 1.75 \pm 0.25$ mV. Analyzing the dV/dI PCAR spectra for all PCs results in gap values of $\langle\Delta_L\rangle = 1.8 \pm 0.4$ meV and $\langle\Delta_S\rangle = 1.0 \pm 0.2$ meV for the large (L) and small (S) gap, respectively, leading to reduced gap values of $2\langle\Delta_L\rangle/k_BT_c = 4.2 \pm 0.9$ and $2\langle\Delta_S\rangle/k_BT_c = 2.3 \pm 0.5$ (see Table II). Here, we used an averaged $T_c \approx 10$ K value obtained by fitting of the temperature dependence of the gap by a BCS-like curve. The calculated average gap value $\Delta_{ave} = (1-w)\Delta_L + w\Delta_S$ resulting from the fit of dV/dI curves at 3 K for all PCs is between 1.3 and 1.9 meV. In this case, the averaged value for all PCs is $\langle\Delta_{ave}\rangle = 1.6 \pm 0.3$ meV. As a result, we received an averaged ratio $2\langle\Delta_{ave}\rangle/k_BT_c = 3.7 \pm 0.7$, which is only a bit higher than the BCS value 3.52.

TABLE II. Averaged data after analyzing dV/dI at 3 K for 25 soft PCs within the BTK model: $V_{\min}$ is the minimum position in dV/dI, $\langle\Delta_{L,S}\rangle$ is the average of large and small gaps, and Z is the "barrier" parameter, $\Delta_{\text{ave}} = (1-w)\Delta_L + w\Delta_S$. T_c is taken equal to 10 K. Note, the data for eight soft PCs created on so-called three-dimensional (3D) FeSe samples are also included in the statistic. The latter resemble bulky pieces, contrary to the usual plate-shaped FeSe flakes.

$\langle V_{\min}\rangle$(meV)	$\langle\Delta_L\rangle$(meV)	$\langle\Delta_S\rangle$(meV)	Z	w	$\langle\Delta_{\text{ave}}\rangle$(meV)	$2\langle\Delta_L\rangle/k_BT_c$	$2\langle\Delta_S\rangle/k_BT_c$	$2\langle\Delta_{\text{ave}}\rangle/k_BT_c$
1.75 ± 0.25	1.8 ± 0.4	1.0 ± 0.2	0.7 ± 0.1	0.17 ± 0.13	1.6 ± 0.3	4.2 ± 0.9	2.3 ± 0.5	3.7 ± 0.7

Please note the Z value (see Table I), which has a low spreading and concentrates around 0.7. The low dispersion of Z testifies in favor of some natural barrier, probably of a semiconducting origin [24]. Besides, as it is seen from Fig. 2(b), Z slightly decreases with temperature, as expected in the case of low barrier heights. Therefore, if it is really a natural barrier, then our assumption $Z_L = Z_S$ in the fit procedure is justified. However, why Z decreases in a magnetic field (see Fig. 3) is not yet understood.

According to the latest data from Ref. [25], where the authors used subkelvin Bogoliubov quasiparticle interference (BQPI) imaging, "the maximum gaps were assigned to each band based on the energy evolution of BQPI to the energy limit $E \to 2.3$ meV for the α band and $E \to 1.5$ meV for the ε band". These values are larger comparing to our data for the large and small gaps. At the same time, the authors of Ref. [25] found an extraordinarily anisotropic ($\Delta_\alpha^{\max}/\Delta_\alpha^{\min} \gtrsim 15$) C_2-symmetric energy-gap structure. Apparently, our data for the large and small gaps represent the averaged gap for the corresponding α and ε bands.

Now, we turn to the antisymmetric part of dV/dI^{as}, which is shown for some PCs in Fig. 1(b). We related the asymmetry of the dV/dI characteristics to thermopower effects in the case of heterocontacts in the thermal regime [26]. In this case, the dV/dI^{as} is proportional to the difference between the Seebeck coefficients $S(T)$ of the contacting materials [27]. Such a correspondence between dV/dI^{as} and the Seebeck coefficient $S(T)$ was observed for PC measurements on [1111] and [122] iron-based superconductors (see Refs. [28,29]). Additionally, we reported such a correlation also for FeSe in Ref. [14]. Our soft PCs mainly had a negative value of dV/dI^{as} and only about one third of all PCs exhibited a positive dV/dI^{as} prior to the dV/dI^{as} sign changes [see Fig. 1(b)]. Here, we must pay attention to the fact that the Seebeck coefficient $S(T)$ in FeSe measured by different authors varies in value, shape, and sign [see, e.g., the inset in Fig. 1(b)]. In addition, $S(T)$ of FeSe polycrystals measured in Ref. [18] is even positive for temperatures up to 500 K. We see in the inset of Fig. 1(b) a remarkable difference in $S(T)$ between single crystals and polycrystals as well as a different sign of $S(T)$ for two polycrystals. This can be the reason for such a variety of dV/dI^{as} for different PCs. Taking into account the huge anisotropy of resistivity of FeSe according to Ref. [21], it is not excluded that thermopower measured along the c direction can have also different behaviors and signs.

V. CONCLUSION

We investigated SC gaps in FeSe single crystals using soft PCAR spectroscopy. We measured dV/dI with the characteristic for the PCAR double-minimum structure versus temperature and magnetic field for about 25 PCs. Analysis of dV/dI data by the extended two-gap BTK model allows one to extract the temperature and magnetic field dependence of the SC gaps. The temperature dependence of both gaps is close to the standard BCS behavior. The PCAR double-minimum structure gradually decreases in magnetic field. Nevertheless, the position of the minima has a weak field dependence, leading to almost field-independent SC gap values. This observation is still not completely understood. Analysis of dV/dI PCAR spectra for all PCs results in gap values of $\langle\Delta_L\rangle = 1.8 \pm 0.4$ meV and $\langle\Delta_S\rangle = 1.0 \pm 0.2$ meV for the large (L) and small (S) gap, respectively, which leads to the reduced gap values of $2\langle\Delta_L\rangle/k_BT_c = 4.2 \pm 0.9$ and $2\langle\Delta_L\rangle/k_BT_c = 2.3 \pm 0.5$. At the same time, the small gap contribution to the spectra is somewhere within 10%–20%. Additionally, the averaged gap value $\Delta_{\text{ave}} = (1-w)\Delta_L + w\Delta_S$ for all PCs amounts to 1.6 ± 0.3 meV, so that the averaged ratio is $2\langle\Delta_{\text{ave}}\rangle/k_BT_c = 3.7 \pm 0.7$, only a bit higher than the BCS value of 3.52. No features in dV/dI spectra that testify to the presence of a gapless superconductivity or the presence of a gap smaller than extracted from the analysis were observed.

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