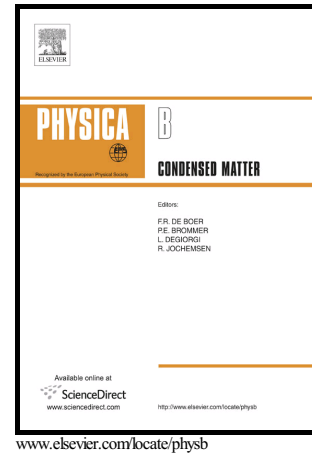


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# Unconventional superconductivity in the strong-coupling limit for the heavy fermion system CeCoIn<sub>5</sub>

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## Abstract

We present scanning tunneling spectroscopy measurements of the local quasiparticles' excitation spectra of the heavy fermion CeCoIn<sub>5</sub> between 440 mK and 3 K in samples with a bulk  $T_c = 2.25$  K. The spectral shape of our low-temperature tunneling data, quite textbook nodal- $\Delta$  conductance, allow us to confidently fit the spectra with a d-wave density of states considering also a shortening of quasiparticles' lifetime term  $\Gamma$ . The  $\Delta(0)$  value obtained from the fits yields a BCS ratio  $2\Delta/kT_c = 7.73$  suggesting that CeCoIn<sub>5</sub> is an unconventional superconductor in the strong coupling limit. The fits also reveal that the height of coherence peaks in CeCoIn<sub>5</sub> is reduced with respect to a pure BCS spectra and therefore the coupling of quasiparticles with spin excitations should play a relevant role. The tunneling conductance shows a depletion at energies smaller than  $\Delta$  for temperatures larger than the bulk  $T_c$ , giving further support to the existence of a pseudogap phase that in our samples span up to  $T^* \sim 1.2T_c$ . The phenomenological scaling of the pseudogap temperature observed in various families of cuprates,  $2\Delta/kT^* \sim 4.3$ , is not fulfilled in our measurements. This suggests that in CeCoIn<sub>5</sub> the strong magnetic fluctuations might conspire to close the local superconducting gap at a smaller pseudogap temperature-scale than in cuprates.

## Keywords:

heavy fermion superconductors, scanning tunneling spectroscopy, superconducting phase, pseudogap phase

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## 1. Introduction

Heavy fermion systems are interesting candidates to study the microscopic coupling giving rise to unconventional superconductivity in concomitance with magnetic order. [1] In particular, CeCoIn<sub>5</sub> is a paradigmatic heavy fermion in which magnetic and superconducting orders seem to cooperate in a phase region close to the upper critical field. [2] The sub-meV value of the superconducting gap in this material renders the task of decoding specific information on the pairing mechanism from the k-dependence of  $\Delta$  a non-trivial one. Nevertheless, there is evidence that the superconducting gap quite likely presents nodes. [3] Although CeCoIn<sub>5</sub> has been one of the most extensively studied heavy fermion superconductors, there are still several open questions, particularly on the role played by the strong antiferromagnetic spin fluctuations [4] for the superconducting pairing glue, as well as the possible persistence of superconducting fluctuations at  $T > T_c$ . [5]

In order to properly answer these questions, measurements on the quasiparticles' excitation spectrum with sub-meV resolution in energy are mandatory. This information can be accessed at the local scale and in real space by means of low-temperature scanning tunneling spectroscopy (STS). However, in CeCoIn<sub>5</sub> this sort of data do not abound. Three previous STS studies claim nodal heavy fermion superconductivity in CeCoIn<sub>5</sub> from either fitting the quasiparticles' excitation spectrum [5], or inferring information on the k-space structure of  $\Delta$ , [6] or detecting close to surface atomic steps a spatial-evolution of low-energy excitations that is typical of nodal superconductivity. [7]. Nevertheless, performing multi-parameter modeling of the conductance depletion in order to extract spectroscopic

information on the superconducting condensate from these experimentally rounded [5] or particle-hole asymmetric [6, 7] STS spectra is a strategy that can be further improved in order to produce more confident results. In this work we measured low-temperature STS spectra of CeCoIn<sub>5</sub> with well defined features in the quasiparticle excitation spectra that are the signature of nodal superconductivity, mounted on a featureless background. The unprecedented d-wave textbook quality of our spectra allow us to infer confident information on the temperature evolution of  $\Delta$  and the quasiparticle lifetime shortening by fitting with a d-wave BCS model. The small discrepancy between our spectra and these fits allow us to suggest the consideration of a fitting model including the strong coupling to spin fluctuations known to affect the shape of STS spectra even in cuprate [8] and pnictide [9] superconductors.

## 2. Experimental

The local quasiparticles' excitation spectra for different temperatures was obtained from STS measurements performed at low-temperatures using a homemade scanning tunneling microscope inserted in a sorption-pump based <sup>3</sup>He fridge (Janis SSV cryomagnetic system). This cryogenic system has a base temperature of 300 mK and a maximum holding time of 12 hours. The atomically sharp Au tip prepared in situ served as one electrode while the sample was the ground electrode. More details on the experimental setup can be found in Ref. [10]. Samples were cleaved at room temperature with a scalpel and immediately cooled down to low temperature. Topographic measurements in such cleaved surfaces show atomically flat terraces of more than  $30 \times 30 \text{ nm}^2$  with steps between terraces of  $\sim 3$  c-axis unit cells ( $7.56 \text{ \AA}$ ) or larger, see for instance Fig. 1. Atomic resolution was not pursued since the STM electronics was adjusted in order to perform measurements of average spectra in nanometer-range fields of view as a function of temperature.

We studied several CeCoIn<sub>5</sub> single crystals from the same batch grown from an indium flux starting from high-purity elements as described in Ref. [11]. Platelet-like single-crystals with the c-axis oriented perpendicular to the flat surface were obtained after extracting the samples from the melt by using a centrifuge. The stoichiometry and crystal structure were confirmed by means of energy-dispersive X-ray spectroscopy and X-ray diffraction. Energy-dispersive spectroscopy scans revealed a good homogeneity throughout the crystals and no traces of spurious phases were detected. The X-ray diffraction rocking curves at different crystallographic directions present a full-width at half-maximum of less than 0.1 degrees.

The high-quality of the samples was further revealed by resistivity, magnetization, thermal expansion and heat capacity measurements. The critical temperature of the studied samples is of 2.25 K as detected by bulk techniques. For instance, Fig. 2 (a) shows the temperature-evolution of the ac-measured heat capacity [12] at 0 and 5.5 T applied along the c-axis. At zero field, on cooling from the normal state the  $C/T$  signal jumps suddenly (in less than 40 mK) at 2.25 K towards the low-temperature superconducting state. We also include for comparison zero-field  $C/T$  data from samples grown by other group. [13] The field-cooling and zero-field-cooling branches of low-field magnetization measurements split at roughly the same transition temperature, see Fig. 2 (b). Field and temperature-dependent ac heat capacity studies [14] yielded the upper critical field  $H_{c2}$  shown in Fig. 2 (c). These results are in agreement with data found in the literature for other CeCoIn<sub>5</sub> samples. [15]

## 3. Results and discussion

We infer the spectrum of quasiparticles' excitation from the superconducting condensate via differential tunnel conductance as a function of bias voltage measurements,  $\sigma(V_{\text{bias}})$ . We perform experiments as a function of temperature in the range of 0.44 to 2.8 K keeping the tunnel junction impedance constant. The regulation tunnel current and voltage were set at  $I_{\text{R}} = 0.3 \text{ nA}$  and  $V_{\text{R}} = 10 \text{ mV}$ , giving a tunnel junction impedance  $\sigma_{\text{R}} = 30 \text{ nS}$ . These parameters were chosen after verifying that they were within the range of an exponential dependence of the tunnel current  $I_{\text{t}}$  with the tip-to-sample distance. The conductance data were obtained by deriving the measured tunnel current as a function of  $V_{\text{bias}}$ , namely  $\sigma = dI_{\text{t}}/dV_{\text{bias}}$ . Figure 3 (a) shows average  $\sigma$  curves at different temperatures with the average taken over local spectrum measured in fields of view in the nanometer range. At all temperatures, the local spectra have good spatial reproducibility.

The low temperature  $\sigma$  curves are remarkably V-shaped at low bias and present a small zero bias conductance (ZBC). In addition, coherence peaks are significantly developed and particle-hole symmetric. Spectra saturate at unity

at energies larger than those of the coherence peaks when normalized by  $\sigma_R$ , indicating that the  $\sigma$  signal measured at low temperatures comes mostly from a superconducting channel of conduction and therefore  $\sigma/\sigma_R$  is a reasonable estimation of the quasiparticle's excitation spectrum. All these features of the spectra are clear fingerprints of a nodal BCS superconducting density of states.

The spectral shape of our data contrasts with the data published in the literature in which these signatures are more elusive. For instance, in the first report on STS data on CeCoIn<sub>5</sub> [5] the tunneling conductance spectra are much more rounded, zero bias conductance is significantly larger (roughly 10 times larger at 0.5 K), and coherence peaks are fainter in comparison to our data. Following works [6, 7] report measurements at  $\sim 0.25$  K and show STS spectra with rounded coherence peaks and a zero-bias conductance of around 40%. In addition, in the  $V_{\text{bias}}$  range that we measure, the spectra shown in Refs. [6, 7] are particle-hole asymmetric and  $\sigma$  does not saturate at  $V_{\text{bias}} > 2$  mV. The latter suggests that in those works the STS spectra data might be strongly affected by the conduction in the heavy bands.

On warming from 0.44 K the ZBC rises at an almost linear rate, indicating that the low-energy excitations are enhancing, see Fig. 3 (b). The coherence peaks gradually faint on warming. Strikingly, at the bulk critical temperature the ZBC does not saturate to the high-temperature normal state value. Indeed, a close inspection to the spectra of Fig. 3 (a) reveals that at 2.4 K (see red arrow), a temperature slightly larger than the bulk  $T_c$ , there is still a depletion in  $\sigma$  at low energies. This depletion persists at larger temperatures and fills in completely at 2.8 K within our experimental noise. Superconducting quasiparticle excitations are still detected at a local scale and within the same energy-scale in a range of temperatures above  $T_c$ . Therefore our data confirm the existence of a pseudogap phase up to  $T^* = 1.2T_c$ . The spanning of the pseudogap phase in our samples is smaller than previously reported ( $T^* = 1.5T_c$  in Ref. [5]).

The textbook-like nodal nature of our spectra supports that fitting the data with a d-wave BCS density of states is a quantitatively sound analysis. We therefore fit the normalized conductance with a model considering a superconducting  $N(E)$  and a constant normal channel  $\sigma_N$  of conductance,  $\sigma/\sigma_R = f'(T) \cdot [N(E) + \sigma_N]$ , with  $f'(T)$  the temperature smearing term equal to the derivative of the Fermi function. The superconducting density of states  $N(E) = \text{Re}[\int_0^{2\pi} d\varphi (E - i\Gamma/2\pi \sqrt{(E - i\Gamma)^2 - \Delta \cos^2 2\varphi})]$  takes into account the Dynes term  $\Gamma$  associated to the shortening of quasiparticles' lifetime and a d-wave  $\Delta$  presenting nodes at the reciprocal space angles  $\varphi = 1, 3, 5$  and 7 times  $\pi/4$ . The only input parameter in the fits is the measurement temperature and the output parameters are the superconducting gap maximum  $\Delta$ ,  $\Gamma$  and a possible constant channel contribution (considered only for formality).

Figure 4 (a) shows the example of fitting the spectra measured at 0.44 K with two different protocols. In the first method, the spectra are fitted considering all the measured points in the whole  $V_{\text{bias}}$  range with the same statistical weight. This fit, shown in blue, yielded a respectable value of quasiparticle lifetime shortening  $\Gamma \sim 0.07\Delta$  with  $\Delta = 0.735$  meV and a negligible contribution of  $\sigma_N = 10^{-14}$ . As a consequence of minimizing the overall square distance between all the experimental data and the model curve, the fit significantly overestimates the ZBC and the coherence peaks' height. The value of  $\Delta$  is in a course way determined by the energy location of the coherence peaks, whereas  $\Gamma$  affects significantly the ZBC and the height and width of the peaks. Increasing  $\Gamma$  lowers the coherence peaks but the result of the fit will not be better: at the same time it broadens the peaks much beyond the measured outer flanks and increases the ZBC even more. On the other hand, decreasing the value of  $\Gamma$  matches better the low-energy V-shaped part of the spectra, but increases too much the height of coherence peaks. On increasing temperature these effects are reduced and for  $T > 1.6$  K the fit matches pretty well the experimental points even at zero bias and at the peaks.

When applying the second analysis method, the spectra are fitted considering the data for  $V_{\text{bias}}$  smaller than the coherence peaks with ten times the weight of the other points. The result of this fitting procedure is shown in black: the low-energy excitations part is perfectly matched by the fit at expenses of magnifying the height of the coherence peaks. The result of this fitting protocol yields a value of  $\Gamma$  three orders of magnitude smaller and a discrepancy with the  $\Delta$  values found with the first method of 6% at maximum for the temperatures studied. For a given thermal smearing the low-energy excitations are only affected by the weight of the superconducting channel and the value of  $\Gamma$ , but the height of the coherence peaks can be modified by the coupling of the excitations with spin collective modes detected in the material at energies closer to  $\Delta$ , as well as by the normal-state band. [16] Therefore, for a better description and fitting of the whole spectral shape of the quasiparticle excitation spectra in CeCoIn<sub>5</sub>, a fitting procedure considering a model that takes into account the coupling with the magnetic susceptibility of the material is required.

Nevertheless, the last approach is beyond the aim of this paper and we will interpret the measured STS spectra

by applying the first fitting protocol (blue curve in Fig. 4 (a)) that tries to match all experimental points with the same weight. In the worst of cases, the differences in the value of  $\Delta$  yielded by both methods is within 6%. The temperature-evolution obtained from the fits for the superconducting gap is shown in Fig. 4 (b). As expected,  $\Delta$  decreases as a function of temperature. The d-wave superconducting gap fitted at low temperatures is smaller than some values previously reported by STS, 0.74 against 0.9 meV at  $\sim 0.5$  K for a sample with  $T_c = 2.3$  K. [5] On the other hand, our value is slightly larger than values reported in another STS work [6] on samples with  $T_c = 2.1$  K. On trespassing  $T_c$  on warming, a finite value of  $\Delta$  is still obtained from the fits up to 2.6 K. Indeed, the depletion of  $\sigma$  at low energies disappears only at  $T = 2.8$  K, see Fig. 3 (a). The temperature evolution of the gap is reasonably well fitted by the BCS dependence for nodal superconductors  $\Delta(T) = \Delta(0) \sqrt{1 - (T/T^*)^3}$ . [17] From the fit we obtain  $\Delta(0) = 0.75$  meV and a characteristic temperature for the local closing of the superconducting gap of  $T^* = 2.85$  K. The latter confirms the plausible existence of a pseudogap phase but in our samples it spans a smaller temperature range  $T_c < T < 1.2T_c = T^*$ , c.f. the  $T^* = 1.5T_c$  value reported in Ref. [6].

The increase of the shortening of quasiparticle's lifetime with temperature roughly mirrors the  $\Delta$  decrease, see Fig. 4 (b):  $\Gamma$  slightly fluctuates around 0.075 meV for  $T < 1.6$  K and for larger temperatures presents a steep increase reaching 0.2 meV at the bulk  $T_c$  and even a value of 70 % that of  $\Delta(0)$  for  $T \sim T^*$ . This evolution reflects the progressive rounding of the low- and coherence peaks-energy regions of the spectra on warming. The shape of the  $\Gamma(T)$  curve also indicates that the shortening of quasiparticles' lifetime increases dramatically on warming within the pseudogap phase. Another magnitude that seems to be in register with the existence of this phase is the constant normal channel of conductance. The insert to Fig. 4 (b) shows that  $\sigma_N$  is negligible at  $T < T_c$  and therefore the superconducting  $N(E)$  term fully accounts for the measured spectra. However, on warming beyond the superconducting phase,  $\sigma_N$  steeply increases up to unity at 2.8 K.

The  $\Delta(0) = 0.75$  meV value obtained from the fit of the  $\Delta(T)$  curve yields a BCS ratio  $2\Delta/kT_c = 7.73$ . Therefore our results also support that superconductivity in the heavy fermion superconductor CeCoIn<sub>5</sub> is in the strong coupling limit. [5] Nevertheless, we disagree with the interpretation stated in Ref. [5] that this phase is reminiscent to the pseudogap of underdoped cuprates. In several families of cuprates, the pseudogap temperature follows a phenomenological scaling  $2\Delta/kT^* = 4.3$ , [18] suggesting that in all these materials the temperature-scale that governs the opening of  $\Delta$  at a local scale is  $T^*$ . In our sample, the value we detect of  $T^*$  is of 2.85 K at best, which gives a ratio  $2\Delta/kT^* = 6$ . Therefore, a larger value of  $T^* \sim 4$  K would be required in order to fulfill the phenomenological pseudogap scaling for the BCS ratio observed in cuprates. This suggests that other microscopic mechanisms in concomitance with magnetic fluctuations in this strong-coupling limit superconductor are conspiring to locally close the gap in the pseudogap phase at a temperature smaller than expected for cuprate superconductors.

#### 4. Conclusions

In summary, the unprecedented clear V-shape and small ZBC measured at low energies, and the prominent coherent peaks detected in our measurements of the quasiparticles' excitation spectrum of CeCoIn<sub>5</sub> at  $T < T_c$  provide more confidence to the interpretation of the data via fits with a d-wave BCS density of states. We therefore present stronger evidence on the d-wave nature of the superconducting gap in CeCoIn<sub>5</sub>. We confirm another previously suggested evidence of unconventional superconductivity in CeCoIn<sub>5</sub>, namely, the existence of a possible pseudogap phase that in our samples would extend up to 1.2 times the bulk  $T_c$ . Since the value we detect of  $T^*$  is of at best 2.85 K, this pseudogap phase does not seem to follow the phenomenological scaling  $2\Delta/kT^*$  found in numerous cuprate families. [18] Therefore the nature of this pseudogap phase in CeCoIn<sub>5</sub> deserves further investigation. The value of  $\Delta(0)$  provides a BCS ratio that suggests that the material is in the strong coupling limit. The latter, as well as the failure to perform a finer matching of the spectra with the proposed simple model, signpost towards the need of including the material spin susceptibility in a strong coupling fitting analysis. [16] This would provide valuable information on the microscopic mechanisms governing the occurrence of unconventional superconductivity in concomitance with magnetic order in heavy fermion superconductors.

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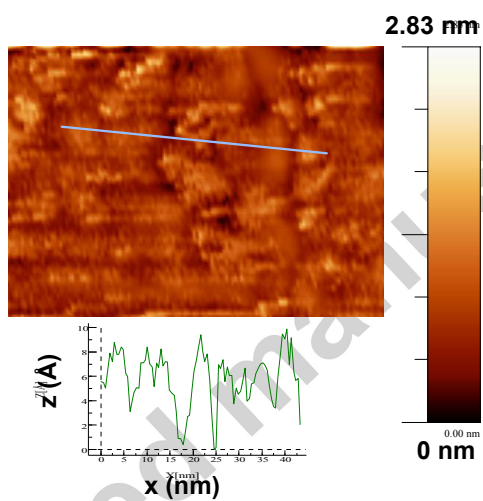


Figure 1: Typical STM topographic image of the CeCoIn<sub>5</sub> single crystals studied. The field of view is of  $55 \times 40 \text{ nm}^2$ . The bottom panel shows a height profile taken along the line shown in the main figure.

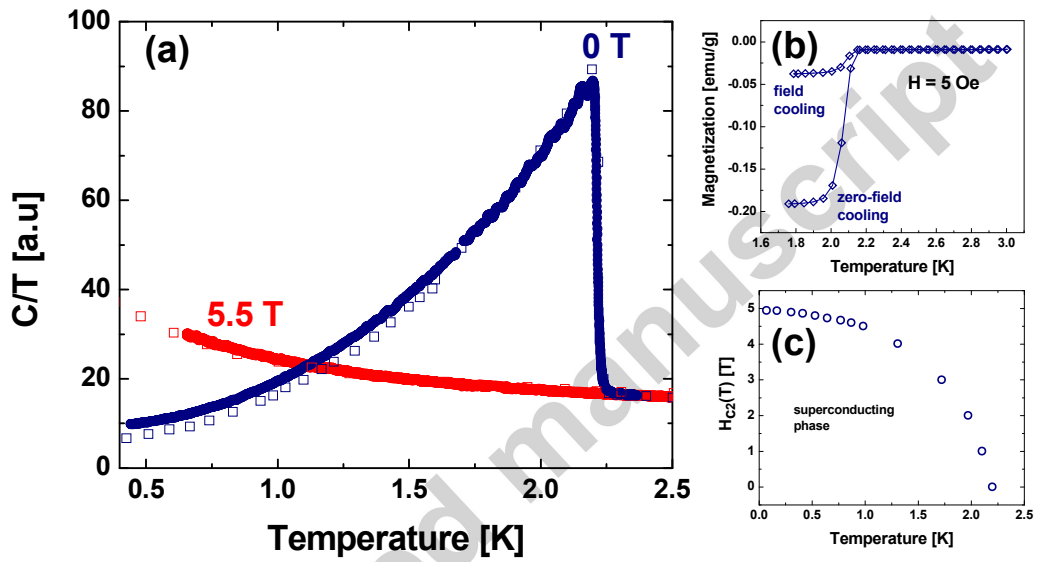


Figure 2: Characterization of the CeCoIn<sub>5</sub> single crystals studied by applying bulk thermodynamic and magnetic techniques. (a) Temperature-evolution of the ac heat capacity at 0 and 5.5 T (normal state data) measured at 0.4 Hz. The transition from the normal to the superconducting state is observed on cooling at 2.25 K. We include dc heat capacity data published in the literature for CeCoIn<sub>5</sub> samples of a different sample grower for comparison (open square points). (b) Field-cooling and zero-field cooling branches of the magnetization of a representative crystal at low fields. (c) Upper critical field  $H_{c2}(T)$  line obtained from temperature and field-dependent ac heat capacity data as for instance shown in (a).



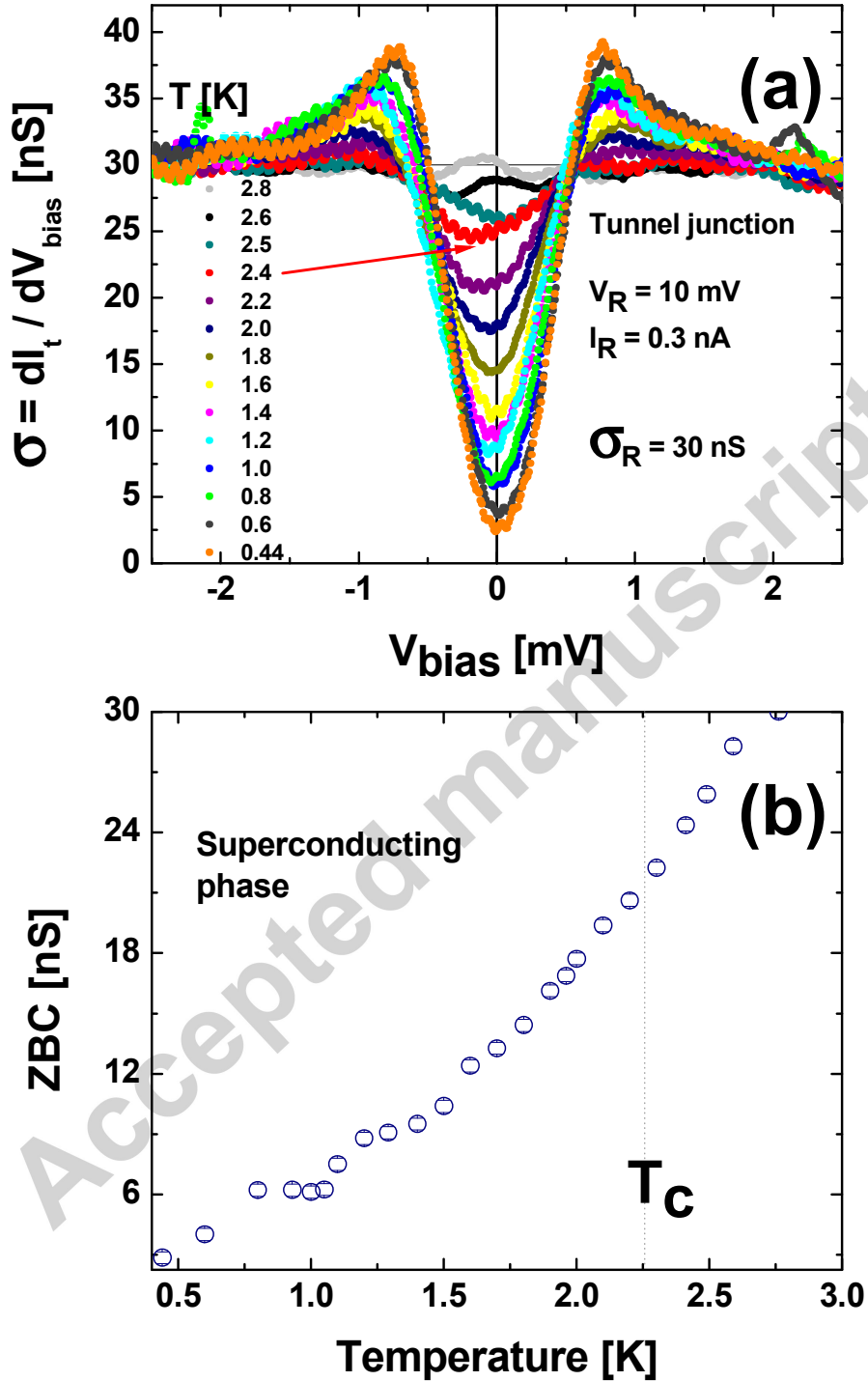


Figure 3: (a) Temperature-evolution of the STS spectra obtained from local tunneling conductance as a function of voltage bias. The temperature is changed between  $0.2$  and  $1.25 T_c$ . The tunnel junction regulation conditions are indicated. The arrow indicates the  $2.4$  K curve measured just above the bulk  $T_c = 2.25$  K. (b) Evolution of the zero-bias conductance as a function of temperature.

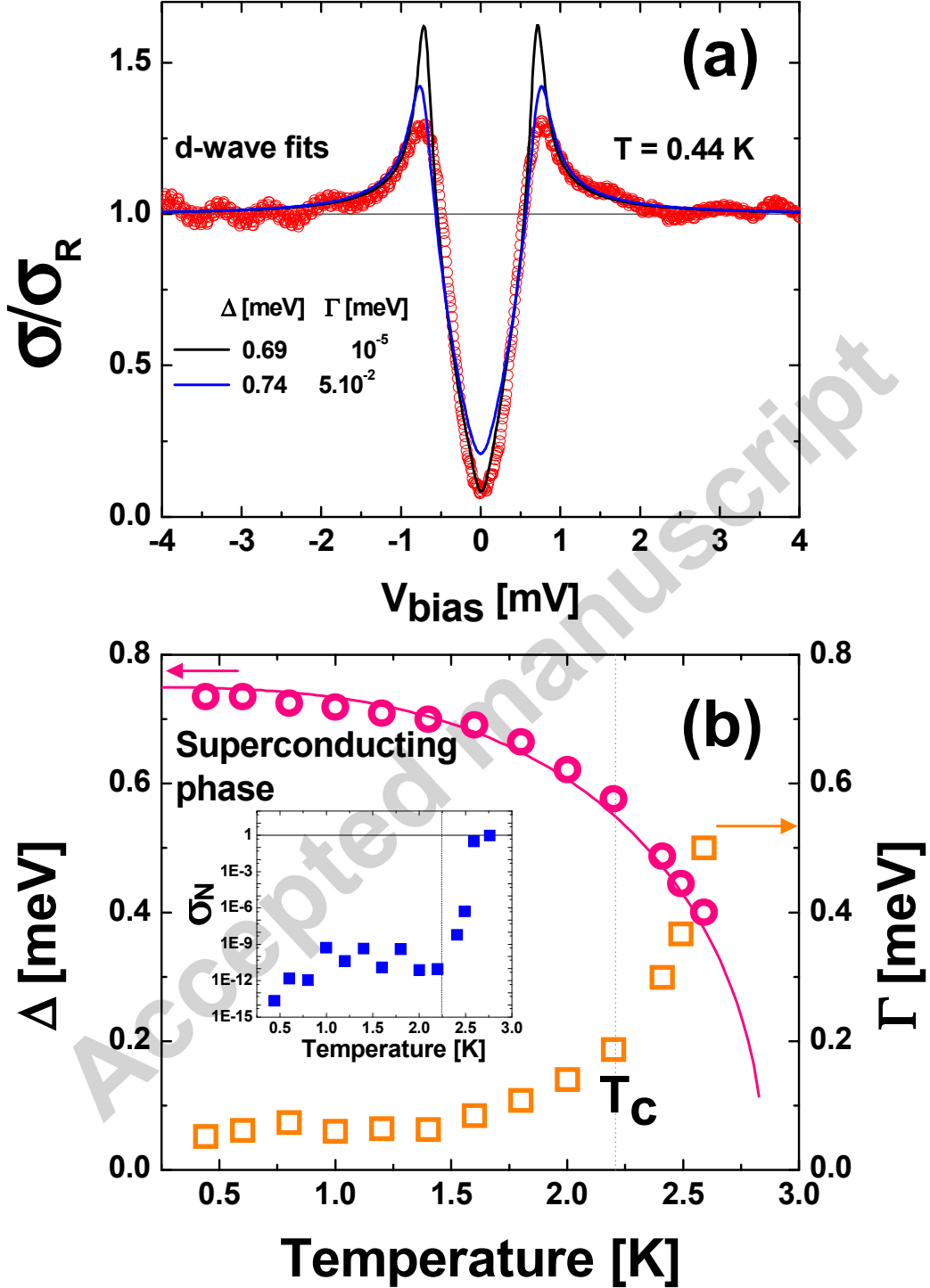


Figure 4: Fitting the quasiparticles' excitation spectrum with a d-wave BCS density of states considering a shortening of quasiparticle's lifetime term,  $\Gamma$ , and a constant channel of conduction,  $\sigma_N$ , see text for further details. (a) Spectra measured at the lowest-studied temperature of 0.44 K (red open circles) and the results of fitting with two different protocols. The blue line results from giving all experimental points the same statistical weight whereas the black one gives 10 times more statistical weight to the sub-gap V-shaped region of the spectra. (b) Temperature-evolution of the superconducting gap  $\Delta$  and  $\Gamma$  obtained from the blue protocol fits. The error in determining these magnitudes is within the size of the points. The bubble-gum-pink line is a fit to the gap data with the nodal BCS gap temperature-evolution  $\Delta(T) = \Delta(0)\sqrt{1 - (T/T^*)^3}$ . Insert: temperature-evolution of the constant normal channel.