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Study of the hidden-order of URu_2Si_2 by point contact tunnel junctions

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Abstract

URu_2Si_2 presents superconductivity at temperatures below 1.5 K and a hidden order (HO) at about 17.5 K. Both electronic phenomena are influenced by Fano and Kondo resonances. At 17.5 K the HO was related in the past to a Peierls distortion that produces an energy gap deformed by the resonances. This order has been studied for more than 20 years and still there is no clear understanding. In this work we studied the electronic characteristics of URu_2Si_2 in a single crystal, with tunneling and metallic point contact spectroscopies. In the superconducting state, we determined the energy gap, which shows the influence of the Fano and Kondo resonances. At temperatures where HO is observed, the tunnel junctions spectra show the influence of the two resonances. Tunnel junction characteristics show that the Fermi surface nesting depends on the crystallographic direction.

Keywords: superconductivity, tunneling and point contact spectroscopy, heavy fermion superconductors

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

1. Introduction

URu_2Si_2, is one of the most studied materials, but after more than 20 years the electronic characteristics are still not well understood. One notable example is the so called hidden order which remains unexplained. Transport measurements in this compound have shown many details related to the general behavior. From room temperature to 70 K URu_2Si_2 presents a characteristic typical of a Kondo system associated with the interaction of heavy f electrons and *spd* conducting electrons. With a Kondo lattice formed at $T_K \sim 70$ K [1, 2], below this temperature the resistivity dramatically drops and at $T = 17.5$ K an anomaly is observed. In the past this was related to a spin density wave (SDW). However this feature is called the Hidden Order (HO). It presents many diverse electronic characteristics that modify other occurring proceses [3–6]. At lower temperatures below ~1.5 K, the compound achieves a superconducting state which many researchers believe must be anomalous in the sense that it does not follow the BCS model [7–12].

As mentioned before, the HO was associated in the past to a Peierls distortion, but careful experiments carried out to detect the Peierls (SDW) distortion did not find any magnetic order, only a quite small staggering magnetism, probably related to magnetic impurities and not related to SDW behavior. This absence implies that the gaping on the Fermi surface may be related to another unknown electronic phenomena [3, 13]. Actually, doubts still persist about the physics behind the anomalies at about 17.5 K. The HO changes transport and thermal properties and opens an energy gap, altering the superconducting behavior [5, 6, 8, 14–17]. The occurrences of the HO and the relationship with the Fano and Kondo resonances is not completely understood. The influence of the two resonances and how it affects the general behavior are not known to date. For instance, the manner that the Kondo lattice at high temperature and the resulting distortion at 17 K modifies the electronic characteristics is not presently understood. Experiments with very sensitive techniques such as tunneling and point contact spectroscopy (PCS) provide the observation of the Fano resonance that distorted the feature of the energy gap, at $T \sim 17.5$ K as observed by Schmidt, et al, with Scanning Tunneling Spectroscopy [18].

Many researchers who investigated this transition demonstrated that this is a second order thermodynamic transition [7, 9, 19–21]. The effects of the resonances at the transition show the consequence of a partial electronic screening that modifies the transport and thermal properties because the strong hybridization between *spd* conducting electrons and heavy localized f electrons [18, 22]. The result of all those processes is that the Kondo and Fano lattices are spatially modulated. The signature of these lattices is displayed as modifications at the Fermi surface. Thus, portions of the Fermi surface at high and low temperatures are distorted and observed with many experimental tests and theoretical studies [1, 4, 7, 23, 24].

This work shows our new experimental observations obtained with techniques that give additional information about the features and characteristics of this compound using tunneling and PCS [25–29].

The studies at high temperatures from 40 to 1.7 K were performed using tunnel junctions to observe the HO that develops at ∼17.5 K. We studied the anomalous features of the gap, using a well characterized URu_2Si_2 single crystal [19, 20, 30]. We measured the spectroscopic characteristics of the crystal in two crystallographic directions.

At lower temperatures, the superconducting state was studied with metallic point contacts (PCS) in the range of temperatures where superconductivity develops. The studies were performed from 2.5 to 0.3 K. PCS junctions were in the diffusive limit but close to the ballistic regime. In all experiments reported here, from 40 K to 0.3 K the influence of the Fano and Kondo resonances was observed. In the superconducting state the influence of both resonances was clearly evidenced. The feature of the energy gap was completely distorted and the effect of these resonances is clearly noted when the evolution with temperature of the energy gap is examined.

In the range of temperatures where HO is set, the influence was studied in two directions of the crystal and the Fano features are different in both directions.

This work reports tunneling and point contact experiments performed in URu_2Si_2 single crystals. At temperatures from 1.7 K to 40 K, tunneling spectroscopy shows the effect of the Fano and Kondo resonances on the energy gap and the dependence on the crystallographic direction. The energy gap in the a direction is bigger than the gap in the c direction. In the superconducting state, with PCS we found that the energy gap as a function of temperature follows the BCS prediction after the Fano anomaly is subtracted from the differential conductance.

2. Experimental details

The URu_2Si_2 single crystals used in this study were grown by the Czochralski method and annealed for one week at 850° C. They have similar characteristics to those reported elsewhere [19, 20, 30]. The crystals have a platelet-like shape with the crystallographic $\boldsymbol{c}$ axis perpendicular to the plane of the platelet. The typical dimension of the crystals used were about $2 \times 3 \times 0.5\,\mathrm{mm}^3$. The samples were characterized by resistance as a function of temperature measurements. The applied current was $100\,\mu$A. The resistance ratio between room temperature and low temperature resistance $R(300\,\mathrm{K})/R(2\,\mathrm{K}) = 38$. The anomaly associated to the HO is observed clearly at about 17.5 K. At lower temperatures, the superconducting transition temperature is at $T_C = 1.37$ K ($R = 0$). This transition is also similar to that observed in other experiments by other researchers. The transition temperature width was 0.15 K. Two type of junctions were formed. Tunnel junctions were built by using a tip of Au(W) wire and the URu_2Si_2 crystal. The insulating barrier that formed the tunnel junction was the native oxide on the surface of the compound and/or the oxide on the tip of the wire. The reason to assume that the junctions in our experiments behave as tunnel or point contacts is due to the oxide formed in the wire or the sample. We have two considerations for this behavior. Firstly, the shape of the differential conductance-bias voltage shows a typical curve of a tunnel junction [31]. In tunnel junctions the zero bias minimum in the curves is displaced out of the origin. This is quite different to a metallic contact junction, where the minimum in the differential conductance always occurs at zero bias voltage [31]. The second important consideration is the value of the differential resistance at zero bias. In our tunnel junctions, this is of the order about 20–50 Ω. Several tries were done until reproducible data was obtained. Junctions with differential resistances at zero bias voltage between 20–50 Ω gave the most reproducible data. The two crystallographic directions of a single crystal were measured with the prepared tunnel junctions, one used to measure over the edge of the crystal axis, the $\boldsymbol{a}$ direction and the other on the plane of the platelet $\boldsymbol{c}$ axis. For the measurements and characterization of the superconducting state, point contact junctions were used. These were formed with the single crystal and a fine tungsten-gold tip with diameter of less than $5\,\mu$m. Those junctions were prepared at room temperature and glued to a glass substrate with Oxford varnish. The area of the junctions was estimated to be $\sim 1\,\mu\mathrm{m}^2$.

The differential resistance $\mathrm{d}V/\mathrm{d}I$ as a function of the bias voltage V of the point contacts was measured with the standard modulation ac lock-in amplifier technique and bridge. The temperature range for characterization of the superconducting state was from 0.325 to about 3 K. For characterization of the hidden order the temperature range was from 1.7 K to 40 K using a MPMS system from Quantum Design as cryostat, whereas below 2 K a ^{3}He Oxford refrigerator was used.

The characterization of the work regime of the point contacts was estimated with Wexler's interpolation formula [32]. We substituted the mean free electronic path $l_i \simeq 100$ Å [30], the resistivity $\rho \sim 40\,\mu\Omega\,\mathrm{cm}$ measured at 2 K [33] and the resistance of the point contacts measured at zero bias voltage. The obtained radii values, between 320 Å and 3700 Å, indicate that the contacts are in the diffuse regime, but not far from the ballistic [34].

An important aspect of the junctions used was the thermal stability. In this work we report studies performed in more than 20 different contact junctions, all shown similar $\mathrm{d}V/\mathrm{d}I(V)$ curves.

3. Results and discussion

Temperature evolution of the differential conductance versus bias voltage from 1.7 to 14.5 K are presented in figure 1.

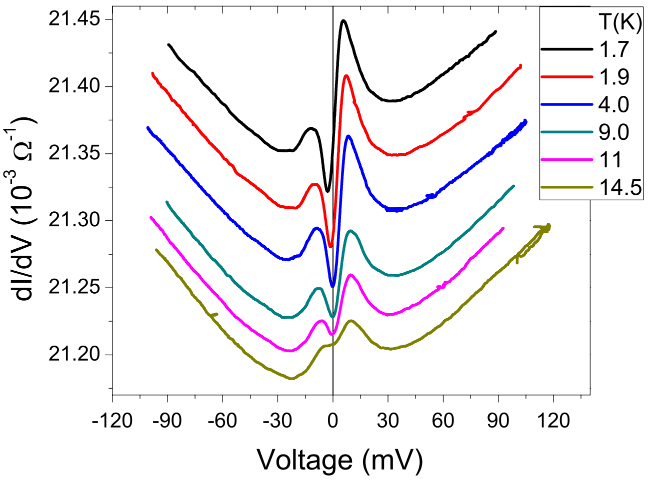

Figure 1. Differential conductance as a function of voltage of URu_2S_2−Au(W) tunnel junction, at temperatures from 1.7 K to 14 K. The differential resistance at zero bias is about ∼46 Ohm. Note the sharp characteristics and the smoothing as the temperature rises. The parabolic background at high bias is typical of a normal tunnel junction. Only the characteristic between ±30 mV presents the Fano resonance, the curves were displaced vertically for better clarity.

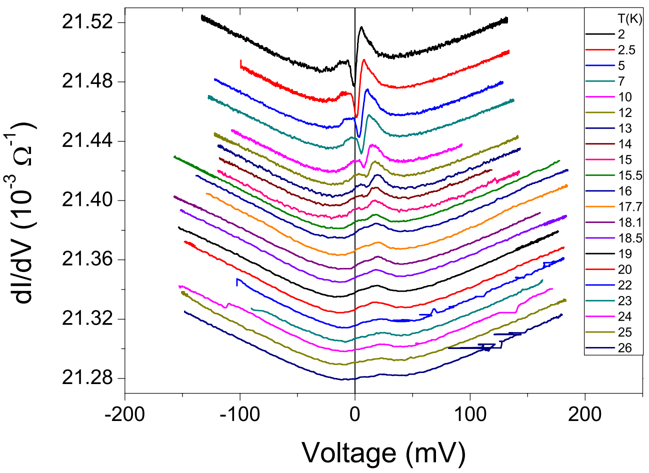

Figure 2. Differential conductance of a tunnel junction in the *c* direction taken from 2 to 26 K. Note the reduction of the predominant sharp characteristics close to the Fermi level when the temperature is increased. The minimum is displaced from the origin to positive bias voltage. With a rise in temperature the structure is smoothed. At high bias voltages, the background of the tunnel junction looks normal. All data was vertically displaced for a clear view.

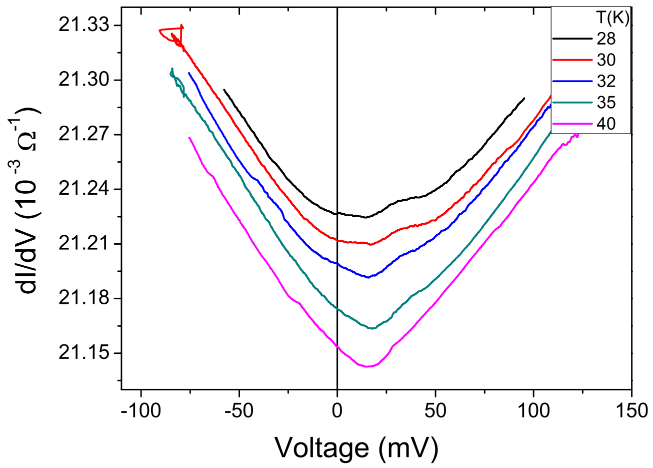

Figure 3. Data for another tunnel junction at high temperature from 28 to 40 K. Here also it is possible to see that small distortions still persist. The minimum now clearly observed in figure 2 is about 18 mV. However, the distortion of the Fano resonance is difficult to see. At 40 K the characteristic is of a typical normal tunnel junction and if some anomalies persist they are difficult to see.

These curves reveal the spectroscopic features of the Fano and Kondo resonances, measured in the *c* crystalline direction. The spectroscopic features of the Fermi surface and the density of states are strongly modified by the Fano and Kondo lattices. Note that in a Peierls distortion the effects are quite different.

The evolution of the spectroscopic features, when temperature is increased, decreases the distortion in the differential conductance. Only close to $T = 17.5$ K where the hidden order develops is the distortion very small. Figure 2 shows measures performed from 1.7 K to 26 K. In *c* direction of the crystal, the distortion decreases as the temperature increases, but it persists at high temperature and the curves measured of the differential conductance characteristic at 1.7 K show a typical Fano resonance shape, with very sharp structure. Those characteristics were also observed with similar detail by Elgazzar, *et al* and Aynajian, *et al* [22, 35]. One set of measurements in the same direction, *c*, up to 40 K, shown in figure 3, displays the typical parabolic background of a normal tunnel junction, with the conductance minimum shifted from the zero bias voltage, out of the origin [36]. However, close examination of the curves still indicates that some features persist at bias voltages of 25–45 mV. Those features may be related to the Kondo lattice.

Figure 3 shows the smoothing of the central peak and the displacement of the minimum out of the origin. However, also the normal parabolic shape of a tunnel junction is noted, but with very small structure [36] perhaps because the remanence of the Kondo lattice. Summarizing, figures 2 and 3 show the effect of temperature on the features around zero bias that we are attributed to the formation of a possible arising of a hybridization gap by the Kondo lattice at temperatures for above 70 K [37]. In figure 4 we show data measured in the *a* crystallographical direction. These data measured from 20 to 2 K. The spectra are different to the measurements in *c* direction. In the *a* direction the differential conductance shows two peaks at ±10 mV, which tend to decrease as the temperature was increased. At 20 K there is no distortion. Note that the Fano anomaly looks completely different to the observed in the *c* direction. The data was measured and was reproducible when measured in different junctions. This Fano distortion is different from the Fano theoretical model [38]. Figure 4 shows two peaks in the differential conductance. These are quite symmetrical with similar magnitude. Those peaks observed at 2 K are separated from the origin at ±10 mV. We may consider that the difference of voltage between the peaks may be the size of an energy 'gap' of the HO, big as the gaping in *c* direction [7]. Similarly as in other figures and directions, figure 4 shows a decreasing structure, tending to be smoothed as temperature is increased. This structure

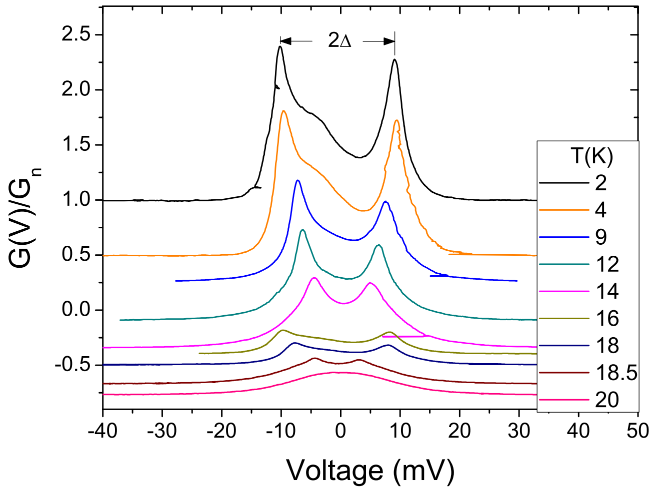

Figure 4. Normalized differential conductance of a tunnel junction measured in the *a* crystallographic direction. These curves show the presence of a bigger distortion, different to the observed in *c* crystallographic direction. Note that the structure of the Fano resonance is distorted with two symmetrical peaks at ±10 mV. The features decrease as the temperature rises. At 14 K, the feature looks similar to a contact junction with a Z parameter about 0.2 [41].

disappears at 20 K. We noted that in ***a*** direction no structure exists at 20 K, but in ***c*** direction it still remains. This is in agreement with the observations by Haule and Kotliar in U or Si site [1]. Also, we have to mention that in total accord with Park *et al* [37] the new features of the Fano resonance, as observed in figure 4 with measurements taken in the *a* crystallographic direction of our sample, clearly may indicate another type of Fano resonance and/or feature of a hybridization gap.

Our experiments show that nesting depends on the crystallographic direction and this may be related to size of the HO gap. Measurements in *a* direction show that $2\Delta = 20$ mV, which is small in *c*, as already determined by Aynajian *et al* as ∼8 mV [22]. With the analysis of the characteristics at temperatures from 40 to 1.7 K, measured in the *a* direction is possible to do a determination of the HO gap; at 2 K the two maxima are ±10 mV and the ratio $2\Delta\ K_B T_C{}^{-1}$ is ∼13.

Point contact experiments reported by Naidyuk *et al* [39, 40], from temperatures above the superconducting transition up to 20 K, show the differential conductance as a function of bias voltage measured in the *c* direction and perpendicular to it. In those experiments the features observed in the *c* direction are not shown, meanwhile in the *a* direction an energy gap is observed with a value about 10 mV, but no features were observed above 17.5 K. Our experiments reported here show features up to 40 K. As mentioned above, this behavior over 17.5 K might be interpreted as the arising of a hybridization gap by normalized bands [37]. Our measurements, in the *c* direction, display more structures which may be associated to an energy gap, persisting above 17.5 K up to 40 K in agreement with Park *et al* [37].

Studies with PCS were performed at low temperature in the superconducting state to see the evolution with temperature of the energy gap. The features were characterized from 3 K to 0.325 K using a ^{3}He as cryostat. In figure 5(*a*) we show the differential conductance between 0.325 K and 1.7 K, all curves were displaced in the vertical direction by a small amount to have a clear view and details. At temperatures above the superconducting state the Fano resonance may be observed as a distortion at the central part of the curves at zero bias voltage. This behavior was observed and analyzed when plotting the evolution of the gap with temperature. At the lower accessible temperature, the superconducting gap feature is well formed. In panel (*b*) of figure 5 we display and plot temperature evolution of the gap.

As shown in figure 5(*b*) the energy gap evolution does not follows the BCS model (see continuous line). This anomalous behavior [20] is the effect of the Fano and Kondo resonances. In figure 5(*c*) the normalized differential conductance of the same PCS with the structure related to the Fano resonance subtracted in all data is presented. The subtracted data was the differential conductance measured at 1.7 K. The data substraction was performed in a similar manner as done by Aynajian *et al* [22]. Figure 5 panel (*d*) presents the results of this procedure. Data of the superconducting gap as a function of temperature now clearly follow BCS. The characteristics shown in figures 5(*b*) and (*d*), show the influence of the resonances in the evolution with temperature of the gap. Our measurements also indicate that the ratio $2\Delta\ K_B T_C{}^{-1}$ is ∼7.86–7.57. with the transition temperature between 1.35–1.4 K. This value is characteristic of a strong coupling superconductor. However, it is very important to mention that recent studies by Kawasaki *et al* and Schemm *et al* [42, 43] have shown time reversal symmetry breaking, using different experimental techniques. Kawasaki, *et al* studied the HO and superconductivity using muon spin relaxation spectroscopy, whereas Schemm, *et al* used the Kerr effect. Both experiments found evidence of broken time reversal symmetry, which has a strong implication for both phenomena occurring in URu_2Si_2. Some features below the superconducting state, performed with polar Kerr experiments were observed and as suggested by Schemm *et al* these effects may imply other physical processes in the superconducting state and well above in the HO phase at 25 K [43]. Therefore, as mentioned by Kawasaki *et al* and Schemm *et al*, those observations may imply a novel pairing mechanism for the superconducting behavior.

4. Conclusions

In summary, we have studied the electronic characteristics of URu_2Si_2 with tunnel and point contact spectroscopies. The spectra show a gap feature with distortions because of Fano and Kondo resonances, at high and low temperatures. At low temperature the superconducting gap has an anomalous evolution with temperature. From 0.3 to 2.4 K, we found if the influence of the Fano and Kondo resonances were subtracted, then the energy gap follows BCS. From 1.7 to 40 K we observed the presence of the resonances that distorted the features on the Fermi surface, those look different in different crystallographic directions. Lastly, in the *c* direction the structure in the differential conductance persists up to 40 K, whereas in the *a* direction it persists up to 20 K. The HO effects are different in both directions of the Fermi surface.

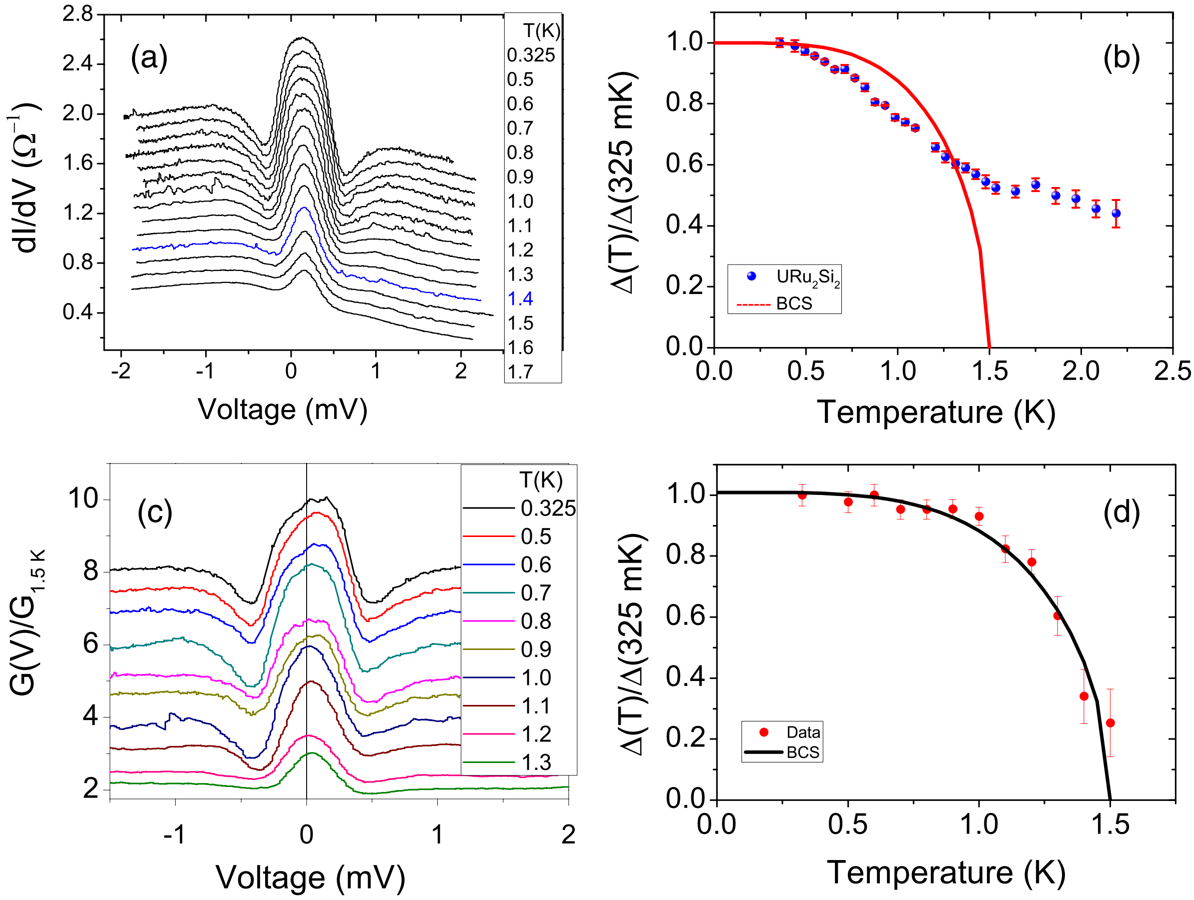

Figure 5. Normalized differential conductance of a point contact in the superconducting state; URu_2Si_2—Au(W). Panel (*a*) shows the rough data measured from 1.7 to 0.3 K. The gap value versus temperature is displayed in (*b*). The evolution does not follow the BCS theory. In order to see the influence of the Fano resonance we subtracted from these data the curve measured at 1.7 K, the resultant curves are shown in panel (*c*), there we plotted the differential conductance normalized at 1.5 K. In (*d*) the superconducting energy gap as a function of normalized temperature is plotted, this data follows the BCS theory quite well. This strong and different evolution with temperature of the gap is due to the Fano resonance.

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