Temperature dependence of the antiferromagnetic state in URu_2Si_2 by point-contact spectroscopy

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Point-contact spectroscopy has been used to study the temperature evolution of the spin-density-wave (SDW) behavior of a URu_2Si_2 single crystal. In the c direction at a temperature of 19 K, we observe the onset of an asymmetric structure in the dV/dI versus V characteristic, and this has been correlated with the development of a SDW gap. We observe that the data follow a BCS behavior. In addition, we have performed tunneling experiments that indicate that only portions of the Fermi surface are being gapped. From these data we have measured the energy gap, $2\Delta = 11.7$ meV, and the ratio $2\Delta/K_BT_N$ which gives a value of the order of 7.2.

Heavy fermion superconducting systems (HF) have many interesting characteristics that are still not very well understood. One of the important aspects that merits careful study is the behavior of the normal state, particularly at temperatures where in some systems a magnetic order begins to build up. The transition from a paramagnetic state to antiferromagnetic order (AF), which in general occurs at low temperatures, is a process that is, also, not entirely understood. Clarifying this transition may provide information about the nature of the correlations and elementary excitations, for example of the spin fluctuations that may be involved in the pairing mechanism of superconductivity.

The heavy fermion superconductor URu_2Si_2 presents the above features; a weak AF transition at about 17.5 K, along with intense spin-wave excitations characteristic of a localized f-electron system in which the ordered moment is approximately $0.04\mu_B$ along the c axis,[1–3] a superconducting transition temperature at around $T_c = 1.2$ K, and Sommerfeld constant of $\gamma = 180$ mJ/mol K^2. The effects of the AF ordering can be readily observed by measurements of resistivity and magnetic susceptibility, and this aspect has been verified by inelastic neutron scattering experiments.[4,5] This AF ordering exists at temperatures well above the superconducting transition temperature, coexists with the superconducting state below the transition temperature, and is a feature present in many heavy fermion systems. The interplay between these two physical processes, which may compete for similar portions of the Fermi surface, is also a mechanism that is not clearly understood. Indeed this may be one of the reasons why the symmetry of the superconducting ground state for heavy fermions is different than that of the BCS superconductors.[6]

An interesting aspect of the HF systems is that the AF arises through the nesting of parts of the Fermi surface. This transition to a new kind of condensate lowers the energy ground state of the system as this approaches its ground state, and opens an energy gap around the Fermi level.[7] In theory, the spin-density-wave (SDW) system must evolve in temperature in exactly the same way as does the condensate of the superconducting BCS system.[8]

Point-contact spectroscopy has been used to probe some of the scattering processes that occur in metals and also in heavy fermions. In particular in the past it was used to investigate both the antiferromagnetic and superconducting states of URu_2Si_2.[9–11] In these studies, the authors related the strong anomalies observed in the characteristic curves of their point contacts as due to the antiferromagnetic behavior below T_N. Hasselbach *et al.*[10] related these features to a gap that develops as the temperature is lowered below T_N. In this paper we report measurements of the differential resistance versus voltage of point contacts and tunnel junctions, made in the c direction of a single crystal of the heavy fermion system URu_2Si_2.

The high quality single crystals used in these experiments were grown in a Czochralski triarc furnace and annealed at 850 °C; this process has been described elsewhere.[12] For the single crystal used in this experiment we measured the superconducting transition temperature $T_c = 1.37$ K, together with the width of the transition $\Delta T = 0.15$ K, and these results confirm the high quality of the crystal. The point contacts were made by placing fine gold wires with sharpened tips on the crystal along the c crystallographic direction. The differential resistance $R = dV/dI$ versus the applied bias voltage V was measured in the range of ± 100 mV, at temperatures in the range 1.7 K$\leq T \leq$ 35 K, using conventional modulation and lockin techniques. The measurements with the point contacts gave differential resistances at zero bias, $R_0 = (dV/dI)_{V=0}$, of the order of 20 Ω to 60 Ω. From considerations of the Knudsen ratio $K = l/d$, where l is the electronic mean free path and d is the radius of the contact, our point contacts are in the regime $K \approx 1$–2,

and can be considered to be within the Knudsen limit.[13] Determination of the radii of the contacts was made using Sharvin's formula plus a Maxwell term multiplied by the function $\Gamma(K)$; this last is a slowly varying function of the Knudsen number. We employed an interpolation formula according to Wexler;[14] $R_0 = (4\rho l/3\pi d^2) + \Gamma(K)\rho/2d$, where ρ is the resistivity of the single crystal taken at low temperature and is of the order of 40 $\mu\Omega$ cm, $l \approx 100$ Å ,[12] and in our case $\Gamma(K \approx 1) \approx 0.9$. Hence, from this we found that the radii of the contacts were in the range 40 Å$\leq d \leq$ 120 Å.

Reproducible nonsymmetric spectra were obtained in the c direction of the single crystal, and these features we assume are due to the scattering of electrons (quasiparticles) by the presence, and evolution with temperature, of the SDW condensate in analogy with the Andreev reflection in superconductors. Kasatkin and Pashitskiĭ (KP)[15] have indicated that such phenomena can occur in systems that present charge-density-wave (CDW) condensation. The scattering of quasiparticles must lead to an increase of the zero bias resistance, as was pointed out by KP. The only difference in this type of Andreev reflection with respect to the classical case for normal-superconducting boundaries (NS) is that in N-SDW interfaces the effect is not accompanied by a change in the sign of the charge. In conclusion, the present system resembles the case of NS interfaces so closely that, in fact, the wave function and the spectrum of quasiparticles can be found from equations which are analogous to the Bogoliubov–de Gennes relationships for superconductors. Therefore, following the analysis of Blonder *et al.*,[16] and Artemenko and Volkov[17] we may assume that for a point contact N-SDW, with a barrier strength $Z = 0$, there will exist scattering of quasiparticles due to the presence of the SDW energy gap, and this should be observed in the dV/dI versus V characteristics as two symmetric valleys at positive and negative voltages, but only when the Andreev reflection is isotropic in the k space.

The experimental characteristics of the point contacts, dV/dI versus V, are the result of more than 40 different measurements with different contacts, some of which were found to be unstable with changes in temperature. Nevertheless, the set of measurements that we include here are the stable and reproducible ones. Special care was taken to maintain the mechanical stability of the point contact with the temperature changes; thus we observed that there was no abrupt variation of the point-contact resistance due to changes of the position of the contact because of thermal contractions. From the above we are confident that our data are reproducible.

From our measurements the form of the evolution of the condensate with respect to the temperature can be established, and can be seen to be related to the order parameter of the SDW system, as explained before. In fact, we conclude that the characteristics follow reasonably well a BCS type behavior.

In Fig. 1 we show representative spectra of dV/dI versus the bias voltage V, as measured with gold point contacts in the c direction of a single crystal of URu_2Si_2. Several important features can be observed in the range of temperature between 2 K and 30 K. Initially at 30 K we have a symmetric point-contact characteristic; at about 19 K a maximum which is offset from the zero bias voltage appears and increases as the temperature decreases. This feature may be due to band bending in the interface N-SDW, similar to the processes that normally occur in metal-semiconductor interfaces.[10] However, in this case we assume that the feature is related to anisotropic Andreev reflections due to an imperfect nesting of parts of the Fermi surface, as considered by Huang and Maki.[18] The feature grows with decreasing temperature because the order parameter changes when the temperature is reduced below the SDW transition. Confirmation of this can be readily noted in two ways in Fig. 2 which presents the variation of the zero bias differential resistance of the contact, R_0, versus the temperature. Here we can observe a rapid decrease of R_0 as the temperature approaches 19 K, and the increase of R_0 below 19 K. Both of these indicate an increase of the anisotropic SDW order parameter due to increased scattering by the Andreev reflection. Another feature from Fig. 1 is the appearance of two asymmetric valleys at positive and negative voltages at around 19 K; the valley separation grows in a continuous way as the temperature decreases. It is interesting to mention that, in Fig. 2, the arrow (at $T = 19$ K) marks the onset of the appearance of the valleys observed in Fig. 1. However, the most dramatic change in Fig. 2 occurs at around 17.5 K, which according to neutron experiments[5] corresponds to the transition temperature. The inset in Fig. 2 shows the variation of the resistance of the sample as a function of the temperature; here the arrow indicates the Néel temperature, of the order of 17.7 K, which corresponds to the abrupt change in the slope of the curve often called the "Cr-like anomaly."[19]

From the above we conclude that the two features of

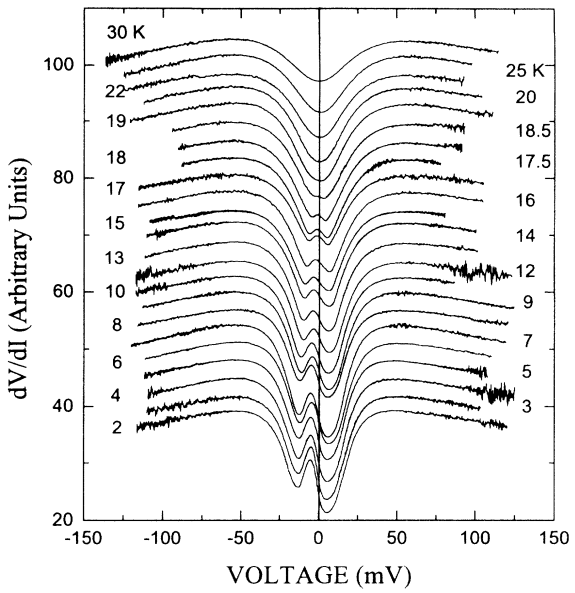

FIG. 1. Differential resistance versus voltage curves of URu_2Si_2-Au point contact, in the c direction; the curves have been displaced vertically by a constant amount of $\sim 2\ \Omega$. The numbers on the curves are the temperatures in K.

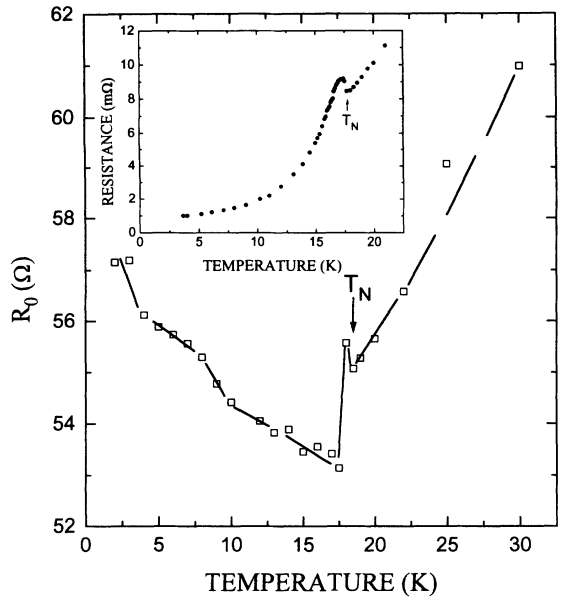

FIG. 2. Differential resistance R_0 versus temperature for a point contact in the c direction. The squares are the experimental values of R_0. The transition temperature T_N is indicated by the arrow, when the SDW gap opens. The increase of R_0 after the transition is due to scattering introduced by the Andreev reflection. The solid line is a guide for the eye. The inset shows the variation of the resistance of our sample of URu_2Si_2 versus the temperature; the arrow indicates that the resistivity transition temperature of the SDW occurs at 17.7 K.

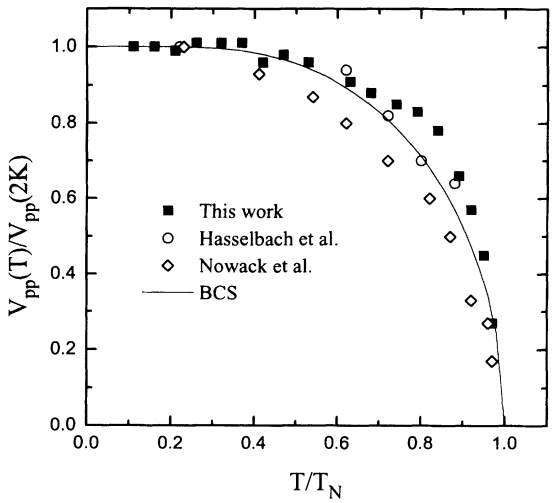

FIG. 3. Black squares are the $V_{pp}(T)/V_{pp}(2\ \mathrm{K})$ versus T/T_N characteristic obtained from data of Fig. 1, of URu_2Si_2-Au point contact, where T_N=19 K. Open circles are data taken from Hasselbach *et al.* (Ref. 10); in this case the normalization temperature was taken as 25 K. Open diamonds are data taken from Nowack *et al.* (Ref. 11), T_N=18.3 K. The solid line is the temperature dependence of the energy gap predicted by the BCS model.

Fig. 1 are the signature of the existence of the energy gap, and are due to the scattering of electrons in an analogous way to the Andreev reflection in superconductors. Accordingly, it is clear that the voltage difference between the valleys, defined as V_{pp}, will be directly related to the energy gap when the system is isotropically nested. Artemenko and Volkov have studied this process, and Huang and Maki[18,20] have considered the case of imperfect nesting, and they conclude that the Fermi surface is gapped only in certain preferential directions. In Fig. 3 we plot the theoretical BCS variation of the energy gap at T, normalized to the gap at zero temperature, as a continuous curve and our data of $V_{pp}(T)/V_{pp}(2\ \mathrm{K})$ as black squares, both versus the normalized temperature T/T_N. $T_N = 19$ K is the transition temperature determined as the temperature at which a maximum starts to develop at zero bias voltage, as can be seen in Fig. 1. It is interesting to note the good agreement of the theoretical model to the experimental points.

It is convenient to compare this work with that of other point-contact experiments on this same material performed by other researchers in the field. At first view, Hasselbach *et al.*[10] obtained data similar to ours (they plot dI/dV vs V characteristics of a point contact of URu_2Si_2-Mo, see Fig. 1 in Ref. 10) but this appears to be in contradiction to our data, as can be seen in the inset of Fig. 1 in Ref. 10. Here they plot the evolution of the amplitude of the structure near zero bias vs the temperature, and the reason for plotting that feature is explained in the text of Ref. 10. Nevertheless, we have measured from their dI/dV vs V curves the voltage difference between the peaks in the same manner as for our data and the results of Hasselbach *et al.* are plotted in Fig. 3 as open circles, versus the normalized temperature (in their case the normalization temperature is 25 K).

Similarly, Nowack *et al.*[11] performed point-contact experiments on URu_2Si_2-Ag using the *ab* or *c* crystallographic directions and they reported the temperature dependence of the normalized characteristic by plotting dV/dI vs V [Fig. 3(a) of Ref. 11]. Essentially they observed the same behavior in both directions. Their experimental data follow a similar trend to that of our data, and they observed the onset of the transition (when the anomalous behavior starts to develop in the dV/dI vs V curves) at 18.3 K. Through the same procedure that we used for the Hasselbach *et al.* data we have extracted the V_{pp} voltage for these data, and it is plotted in our Fig. 3 as open diamonds. We can conclude from this plot that in fact all of the sets of data follow closely the BCS model. Thus, the data sets are essentially consistent and clearly demonstrate that the assumption for the theoretical model and the physics behind the SDW systems are similar to the formation of the condensate in the superconducting BCS system.

Nevertheless, one difference persists among these sets of data and this is that the transition temperature of 19 K that we observe in our point-contact experiments is different from the 25 K observed in Ref. 10, and 18.3 K of the Nowack *et al.* paper.[11] We do not have a complete explanation for this difference, but it is important to remember that point-contact spectroscopy is a very local probe, much more so than many other experimental tech-

niques, e.g., neutron experiments, resistivity vs temperature, and specific heat measurements, and therefore this technique may be more sensitive to small changes in the electronic spectrum due to local imperfections, defects, or stresses in the crystalline material, and apparently this may be what is occurring.

In order to understand the strength of the interaction that gives rise to the formation of the SDW system it is important to know more about the order of magnitude of the energy gap. The most direct way to do this is to perform tunneling experiments. Our tunnel junctions were formed using a single crystal of URu_2Si_2 and a sharp tip of a thin aluminum wire, diameter $\phi = 5$ μm, as the second electrode. We took advantage of the native oxide that forms on aluminum and this was used as the barrier layer. With this type of tunnel junction the area of the junction is ≤ 25 μm^2. The measurements were taken injecting the current along the c direction of the crystal. One of the drawbacks of this type of junction is related to the poor mechanical stability, particularly with changes in temperature. Nonetheless, our tunnel junction experiments are a good complement to our point-contact measurements, since it is possible to demonstrate the existence of the energy gap in a direct manner. Additionally, we were able to measure the strength of the coupling, which is an important aspect that can provide extra information about the type of excitations that play a role in the phenomenon under study.

Figure 4 shows a representative curve of a tunnel junction of the type URu_2Si_2-Al_2O_3-Al taken at 4 K. In the same figure we show the normal state with a broken line,

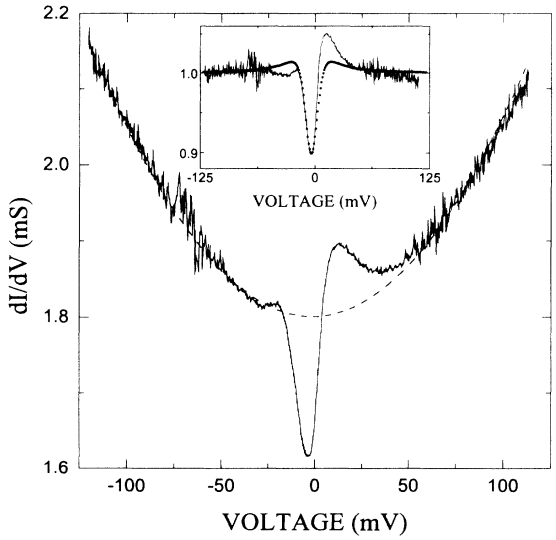

FIG. 4. Tunneling characteristics dI/dV versus V of a URu_2Si_2-Al_2O_3-Al junction at a temperature of 4 K. The dashed line is the normal state taken at 30 K. Notice the change of the curve due to the opening of the energy gap. The inset shows the curve normalized to the normal state at 30 K. The energy gap is $2\Delta = (11.7 \pm 0.2)$ meV. The parameter $\varepsilon_0 = 12$ meV indicates that parts of the Fermi surface remain ungapped in the c direction of the crystal.

at 30 K. Two main features can readily be noted: one that suggests the opening of an energy gap, and the second, the very strong asymmetry with respect to zero voltage with features similar to those found in our point-contact experiments. The energy gap 2Δ was determined using a smeared BCS density of states model, given by $N_{SDW}(E) = \mathrm{Re}\{(E - i\varepsilon_0)/[(E - i\varepsilon_0)^2 - \Delta^2]^{1/2}\}$. In this equation we relate the parameter ε_0 with the number of electronic states that remain unnested after the Fermi surface has gapped in a certain direction of k space, due to the formation of the SDW condensate. The inset in Fig. 4 shows the tunneling spectra normalized to the curve at 30 K, with the smeared density of states $N_{SDW}(E)$ that fits the experimental data plotted as a dotted curve. The values for the energy gap 2Δ and ε_0 are $2\Delta = (11.7 \pm 0.2)$ meV and $\varepsilon_0 = 12$ meV. It should be noted that $\varepsilon_0 \geq \Delta$ gives a clear indication of the gapless nature of this system due to the imperfect nesting.[18] Here it is important to point out that by gapless behavior we mean that the gap only develops along certain directions on the Fermi surface which results in the asymmetry, with respect to the zero bias voltage, of the *gap characteristic* as observed in both our point-contact and tunneling measurements. Thus the large value of the ε_0 parameter that fits the data indicates that only some portions of the Fermi surface are being nested.[18] Maple *et al.*[7] have also reported indications of the gapless nature of the SDW condensate through specific heat measurements. They calculated that only about 40% of the total Fermi surface is used to form the SDW condensate. The strength of the interaction that couples the spins and the elementary excitations, which together form the SDW system, can be obtained from the ratio $2\Delta/K_BT_N$. From the value obtained for the energy gap we find that $2\Delta/K_BT_N = 7.2$, indicative of a very strong coupling interaction. Additionally, measurements of the energy gap in this system have been performed in the past using other spectroscopic techniques, particularly inelastic neutron scattering,[21] specific heat measurements,[7] nuclear magnetic resonance,[22] and far-infrared reflectance.[23] The reported energy gaps vary between 8 and 12 meV depending on the technique used and on the interpretation of the measurements. However, up till now a direct measurement of the energy gap of the URu_2Si_2 system by electron tunneling spectroscopy has not been reported to our knowledge. We believe that the present tunneling experiments clarify the situation concerning the size of the gap, and also the anisotropic nature of the nesting of the Fermi surface.

In conclusion, we summarize the main contributions of this paper. We have performed extensive point-contact measurements on a single crystal of the heavy fermion superconductor URu_2Si_2. The evolution with temperature of the features related to the SDW, observed in the range of temperatures between 2 K and 30 K, is in good agreement with the BCS theory. These features occur in an analogous way to the Andreev reflections in BCS superconductors. According to our data the transition temperature onset is at 19 K. Our tunneling data show that the order parameter related to the SDW is anisotropic and that only certain portions of the Fermi surface are

nested. From these data we have measured the energy gap $2\Delta = 11.7$ meV, and the ratio $2\Delta/K_BT_N = 7.2$.

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