Superconducting and spin-density wave phases probed by scanning tunneling spectroscopy in the organic conductor (TMTSF)₂ClO₄

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Among the various families of materials presenting unconventional superconductivity, organic materials occupy a very noticeable place thanks to their strong quasi-one dimensional electronic properties and close proximity to a quantum critical point [1,2,3]. Of particular interest are the $(TMTSF)_2X$ salts where X is either the centrosymmetric anion PF₆ or the non-centrosymmetric ClO₄. Since a superconducting phase develops at ambient pressure in the ClO₄ compounds while pressure is needed for the PF₆ materials, more studies were carried out on the ClO₄ materials. There are several experimental hints of unconventional superconductivity developing in the (TMTSF)₂ClO₄ compounds. For instance, ⁷⁷Se NMR and field-angle-resolved specific heat measurements are consistent with a spin-singlet superconducting order presenting line nodes [4,5]. Direct tunneling spectroscopy of the quasi-particle density-of-states in the superconducting state would be highly desirable to be confronted to other available experimental results in order to restrict the possible gap symmetries.

In the present work, we have probed the tunneling quasi-particle local density-of-states (LDOS) of (TMTSF)₂ClO₄ single crystals cleaved under ultrahigh vacuum. Taking care in the sample surface preparation, we could both image crystallographic surface planes using scanning tunneling microscopy and probe the local DOS at 0.3 K and 2.1 K in various areas and samples. All samples were cooled down at a speed less than 33 mK/mn, favoring a bulk superconducting phase having a macroscopic $T_c \approx 1.2$ K from ex-situ electronic transport measurements. At the very top surface planes we found fewer superconducting regions than what could be expected from the T_c most probably because the cleavage process induces disorder in the anions surface lattice. In such surface superconducting regions the excitation spectra probed by scanning tunneling spectroscopy (STS) are not consistent with an s-wave order parameter thus advocating that superconductivity is indeed unconventional in (TMTSF)₂X salts [6]. In addition, a strongly V-shaped background is present in the normal state LDOS underlying the strongly correlated quasi-1D character of the LDOS due to quantum criticality that links antiferromagnetism with superconductivity in this material [3]. Finally, local regions could also be found at the surface where STS reveals a U-shape energy dependence, consistent with spin-density wave phases, most probably induced by the local surface disorder in the anions lattice. Overall, our results suggest that at the surface, an inhomogeneous phase has developed that presents similarities with a recent bulk study where the amount of bulk disorder has been tuned by the cooling speed [7].

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