Spatial cross-correlations between local electron-electron interaction effects and local superconducting energy gap in moderately-disordered NbN ultrathin films

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We have studied by scanning tunneling spectroscopy (STS) at very-low temperature the local electronic properties of a superconducting NbN disordered ultrathin film grown ex situ. The NbN film has a $T_c = 3.8$ K about $0.25 T_{C-bulk}$. The STS measurements performed in ultrahigh vacuum at 300 mK show that an inhomogeneous spatial distribution of the superconducting energy gap $\Delta$ has developed in the material. Concomitantly there exists a large depression in the local density-of-states, associated to Altshuler-Aronov effect, which presents strong spatial variations. Our analysis shows that the local Altshuler-Aronov effect is strongly correlated with the local energy gap value. Our results show that a local Finkelstein mechanism is at play and reduces $\Delta$ in moderately disordered NbN superconducting films.

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Non-magnetic disorder was initially thought to have little effect on the superconducting properties of conventional thin films, as far as time-reversal symmetry is preserved [1][2]. Decades of intense experimental and theoretical works have ended to the opposite conclusion showing that beyond a critical disorder all systems transit either to a metallic or to an insulating state [3][5]. For thin films that are not single-crystals [6] but consist in coupled nanocrystals two different classes of systems exist. A qualitatively different superconducting behavior is observed between so-called granular and homogeneous disordered thin films. In granular systems a poor electrical coupling between the nanocrystals makes the films a disordered array of Josephson junctions [5][7][10]. As a consequence, their superconducting properties are controlled by the ones of the individual nanocrystals and their phase relationships. On the other hand, in homogeneous disordered thin films the nanocrystals are much better electrically coupled to each other. Thus their superconducting properties are controlled by the subtle interplay between the precise non-magnetic disorder distribution and electron-electron interaction effects [3][11][13].

In granular thin films the critical temperature $T_c$ and superconducting energy gap $\Delta$ almost do not change upon increasing disorder [5][7][10]. In contrast, in homogeneous systems macroscopic measurements show that $T_c$ and $\Delta$ monotonously decrease with rising disorder corresponding to $k_F l$ decreasing from a value much larger than one toward unity [3][4][11][13] ($k_F$ being the Fermi wavevector and $l$ the elastic electron mean-free path). This effect has been satisfactorily explained by a reduction of the electronic screening upon increasing disorder and is called the “fermionic” mechanism [14][15]. Few superconducting films made of Bi or Pb behave this way and follow the so-called Finkelstein scenario down to very close to the superconductor-metal or superconductor-insulator transition (SIT) [12][16]. Other materials like InO, TiN or NbN show different experimental signatures upon increasing disorder [17]: an increasing emergent granularity develops in the superconducting properties, accompanied by the decoupling of the single-particle gap probed by tunneling from the $T_c$, together with a pseudogap occuring in a significant temperature range above $T_c$ [18][23]. These experimental features have been addressed theoretically by two prototype models based either on a fermionic [21][25] or bosonic approach [1]. The former approach leaves localized fermions in the insulating state while the latter accounts for the presence of localized Cooper pairs.

However reality is richer than these two limiting scenarios because most films present an interplay between “fermionic” and “bosonic” effects. In the litterature there is a trend to believe that for weak to moderate disorder fermionic effects dominate while bosonic ones take over for stronger disorder. Nevertheless there is a lack of experimental data combining macroscopic and local measurements on the same system to explore carefully this interplay.

With this respect, an interesting case is the one of NbN thin films. With increasing disorder they present a continuous reduction of $\Delta$ and $T_c$, in agreement with the Finkelstein picture, accompanied by emerging and increasing gap inhomogeneities as $k_F l$ decreases [19][22][24]. The development of these inhomogeneities together with the emergence of a pseudogap regime above $T_c$ are ini-
FIG. 1: (a) Square electrical resistance of a 2.1 nm thick NbN film grown on sapphire, measured in the same stage where STM is performed. $T_c$ is defined when $R_{\text{square}}$ reaches zero resistance, leading here to $T_c = 3.8 \text{ K}$. (b) Typical local $dI/dV(V)$ spectrum measured at $T = 300 \text{ mK}$ between $[-4.5;+4.5] \text{ mV}$. Set-point for spectroscopy $V = -10 \text{ mV}$, $I = 150 \text{ pA}$. (c) Local $dI/dV(V)$ spectrum measured at the same location and temperature as in (b) but on a larger energy scale $[-28;+28] \text{ mV}$. The large depletion of the LDOS seen around the Fermi level has a characteristic V-shape due to electron-electron interaction effects enhanced by disorder, also called Altshuler-Aronov effect. The red (lighter) curve shows a fit of this V-shape dependence.

Our samples consist of ultrathin NbN films grown ex situ on sapphire substrates. Details of the fabrication process may be found in [27]. Transmission electron microscopy (TEM) measurements show that the films consist of NbN nano-crystals of lateral size 2-5 nm. We selected films having a critical $T_c$ about $0.25T_{c-bulk} = 3.8 \text{ K}$, so that seeming “bosonic” properties like gap inhomogeneities and pseudogap features have already developed. The nominal film thickness, obtained from the evaporator’s calibration, is about 2.1 nm. We studied the local superconducting properties of such as-grown films by scanning tunneling microscopy/spectroscopy (STM/STS) at $300 \text{ mK}$, using PtIr tips, in an ultrahigh vacuum homemade setup. The presented $dI/dV$ measurements were obtained by numerical derivation of single $I(V)$ curves. $T_c$ was extracted from in situ 4-points electrical resistivity measurements performed in the STM stage during the same run as the STM/STS measurements. The temperature dependence of the resistivity showing the superconducting transition is shown in Fig.1.

STM topography measurements show that the film surface is very flat (see Fig.2a). The observed nanoscale structures are consistent with nanocrystals revealed by TEM. In order to probe the superconducting single-particle excitation spectrum with a high-energy resolution, we performed STS measurements at $300 \text{ mK}$. A characteristic $dI/dV$ spectrum measured locally in the range $[-4.5;+4.5] \text{ mV}$ is shown in Fig.1b. It presents a fully gapped LDOS with well-defined superconducting coherence peaks. For energies larger than the ones of the coherence peaks it is seen that instead of recovering a standard normal DOS having a lower amplitude than the height of the coherence peaks (as in the Bardeen-Cooper-Schrieffer, BCS, case), the LDOS increases even above the peaks height. This behavior is typical of systems where electronic correlations combined to disorder reduce the tunneling DOS at the Fermi level ($E_F$) [28, 29], an ef-
FIG. 2: Spatial cross-correlations between superconducting gap variations and electron-electron interactions. Measurements are performed at 300 mK in zero magnetic field. (a): STM image of a 300 × 250 nm$^2$ area of the NbN sample. The height variation $Z$ is in nm. (b): Color-coded map presenting the local superconducting energy gap values $\Delta$ (in meV) measured in area (a) by STS. (c): Color-coded map presenting the local exponent $\alpha$ characterizing the amount of local electron-electron interaction present in area (a). (d): Color-coded map showing the cross-correlation between the topography $Z(x, y)$ shown in (a) and the gap map $\Delta(x, y)$ shown in (b). No cross-correlations are found. (e): Histogram of $\Delta$ values occurring in the map shown in (b). (f): Histogram of $\alpha$ values occurring in the map shown in (c). (g): Color-coded map showing the cross-correlation between the gap map $\Delta(x, y)$ shown in (b) and the exponent map $\alpha(x, y)$ shown in (c). There is evidence for strong spatial cross-correlations: where local electron-electron interactions are stronger (larger $\alpha(x, y)$) $\Delta(x, y)$ is smaller and vice-versa. (h): Representative $dI/dV$ quasiparticle excitation spectra measured by STS in the regions seen in panel (b). Brighter (respectively darker) spectra are measured in brighter (darker) regions of panel (b). (i) Same spectra as in (h) but over an energy scale 20 to 30 times larger than $\Delta$. Brighter (respectively darker) spectra are measured in brighter (darker) regions of panel (c).

In order to better probe and characterize this Altshuler-Aronov effect, Fig. 1 shows a $dI/dV$ spectrum measured on the same location as before but over a [-28;+28] mV energy range, i.e. 20 times larger than $\Delta$. It is now seen that the tunneling LDOS is strongly reduced over an energy range much larger than the superconducting gap. Our analysis shows that outside the superconducting energy gap the measured LDOS follows a simple power-law as a function of the applied bias voltage $V$. A fit to such dependence is presented in Fig. 1. Theoretically such a simple dependence is expected in the framework of dynamical Coulomb blockade theory, when the temperature is much smaller than the effective charging energy [30]. Remarkably, dynamical Coulomb blockade theory has being shown to be formally equivalent to tunneling into a 2D metal with electron-electron interaction effects [31]. We have already used such an equivalence when modeling the superconducting proximity effect between a Pb single nano-crystal and a 2D metallic homogeneously disordered Pb film [32–34].

Here comes the main finding of our work: we found a systematic spatial correlation between the local energy gap value $\Delta$ and the strength of the Altshuler-Aronov effect, quantified by the exponent $\alpha$ characterizing the power law $dI/dV = V^{\alpha}$. At each location of the 300 × 250 nm$^2$ area shown in Fig. 2a, the local gap value was extracted every square nanometer from the peak-to-
peak distance in each $dI/dV$ curve. The extracted map of the gap value is shown in Fig. [2]. The gap energy ranges between 1 and about 1.5 meV and its spatial distribution shows emerging local inhomogeneities, as reported previously in several similar systems [18, 22, 23, 26]. We call supergrain a region of size $l_{sg}$ having a constant gap value. We find $\xi < l_{sg} < 50$ nm, $\xi$ being estimated to be 5-10 nm at 300mK [22]. The histogram of the gap values is shown in panel 2 and is close to a Gaussian distribution (reference A . Larrea Benfatto analyse desitrib de gaps ? ou bien García-Garcia ?). We provide now for the first time a local mapping of the disorder distribution combined to electron interaction effects which are together responsible for the spatial distribution of the gap inhomogeneities.

At each same location where the gap value was measured, we present the local Altshuler-Aronov strength characterized by the value $\alpha$ extracted from a fit of the $dI/dV$ spectrum measured in the larger energy range [5;30] meV. The slight asymmetry existing in our tunneling spectra between positive and negative energies comes probably from a non-constant tip DOS. It leads to slightly different quantitative $\alpha$ values for positive and negative energies. Nevertheless, it does not modify our analysis and conclusions since it just shifts the absolute $\alpha$ values.

The map of the Altshuler-Aronov exponent is presented in panel 2c and the histogram of the exponent values in panel 2d. The cross-correlation between the gap map shown in panel 2b and the exponent map in 2c is presented in panel 2g. A very large anti-correlation of about -0.5 is found. If both the gap map and Altshuler-Aronov exponent map would have been uncorrelated a value between -0.1 and +0.1 should have been found. This implies that in NbN the larger the local energy gap value $\Delta$ is in a supergrain, the smaller is the local Altshuler-Aronov exponent $\alpha$. This corresponds precisely to the situation illustrated on panel 2b showing two representative $dI/dV$ spectra, one having a larger (brighter curve) and smaller (darker curve) gap value linked to a respectively flater (brighter) and steeper (darker) Altshuler-Aronov background in the LDOS (shown in panel 2).

One could naively think that there could exist a direct correspondence between the spatial distribution of the NbN nano-crystals encoded in the STM topography and the gap map or Altshuler-Aronov exponent map. In fact we found no cross-correlation between the local gap values and the topography of the probed area (see panel 2f). This is in agreement with our previous work showing that emergent inhomogeneous regions of size $l_{sg}$ are not in a simple one-to-one correspondence with the spatial distribution of nanoscopic NbN crystallites [26]. In contrast, the present results furnish a new way of characterizing locally and quantitatively the underlying disorder distribution combined to electron interaction effects which lead to the appearance of supergrains of size $l_{sg}$.

We see a demonstration of a very strong spatial cross-correlation between the strength of the local electron-electron interaction effects and the local reduction of the energy gap value. It is very important because theoretical models describing emerging superconducting inhomogeneities in disordered thin films usually neglect electron-electron interaction effects as a source of these inhomogeneities [24, 25, 55] and solely attribute them to disorder effects on the single-particle electronic spectrum. On the other hand in the Finkelstein scenario [13, 15], electron-electron interaction effects are well-known to be responsible for a global macroscopic effect affecting the disordered thin films as a whole : the $T_c$ reduction goes together with the energy gap reduction in the “fermionic” picture, as a consequence of reduced electronic screening. With our results we see that both “fermionic” and “bosonic” pictures are combined in NbN in a local mechanism where the local gap reduction is a consequence of the local strength of the electronic repulsion combined with a particular spatial distribution of the disorder.

Interestingly, we see that former STS studies that have probed either the superconducting energy gap or the height of the coherence peaks in moderate to strongly disordered thin films [21, 23, 39] suffer from a large uncertainty in the precise determination of these values. The raw $dI/dV$ tunneling spectra should be analyzed not by dividing by a guessed background or by a background inferred from the normal state, but by a proper deconvolution with a $V^\alpha$ dependence or a full DCB analysis. It may change significantly some of the results obtained for the most disordered systems where the peak height becomes (very) small. It should also allow a much better quantitative analysis of the experimental order parameter distribution enabling proper comparison with various theoretical models. Our results should thus help shedding new light on the interplay between “fermionic” and “bosonic” pictures to accurately describe weak to moderate disorder effects and emergent granularity in ultra-thin superconducting films [39, 41] not too close to the superconductor-insulator transition.

References

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