Yu-Shiba-Rusinov States and Long-lived Bosonic Excitations in Granular Aluminium

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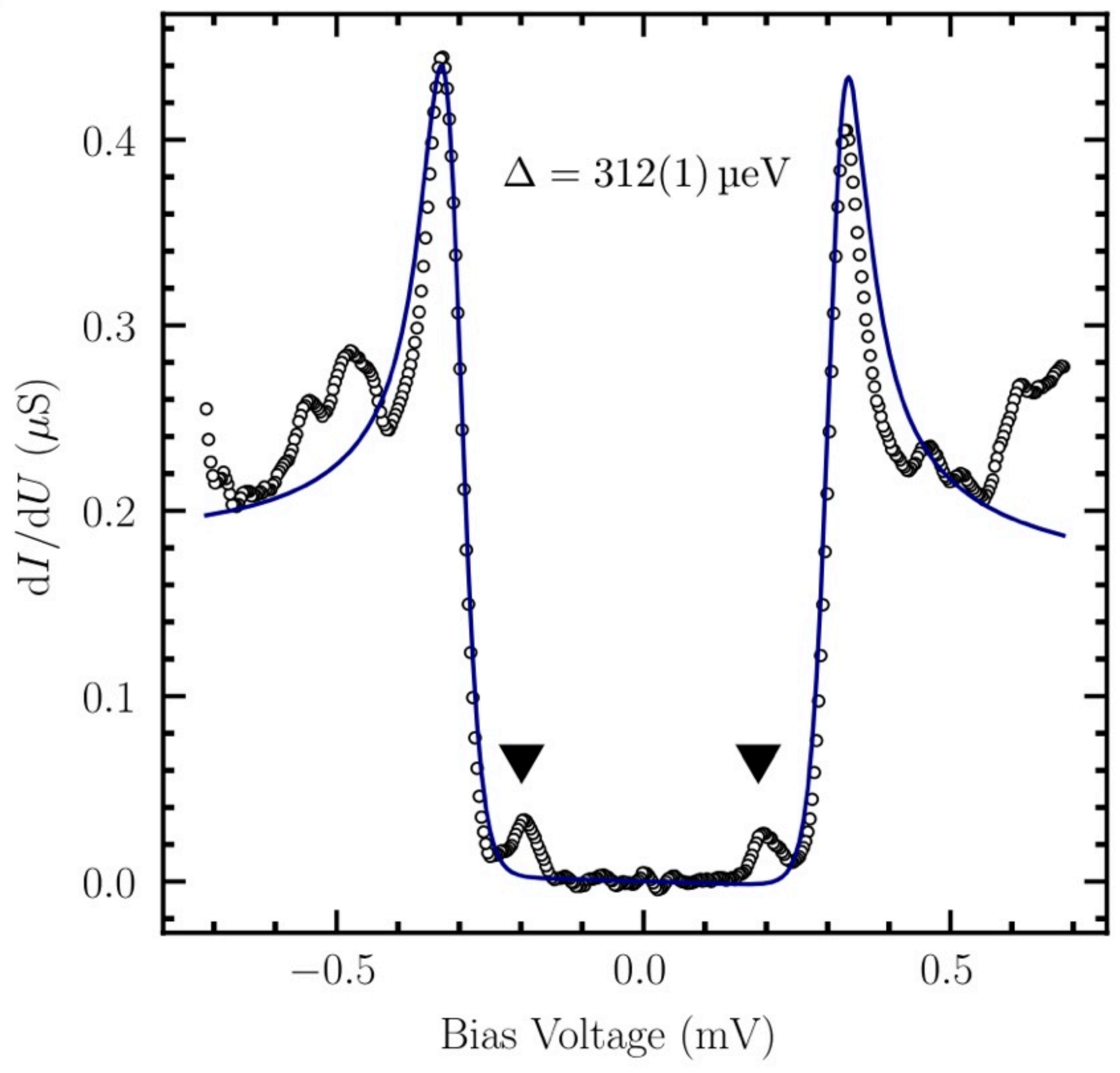

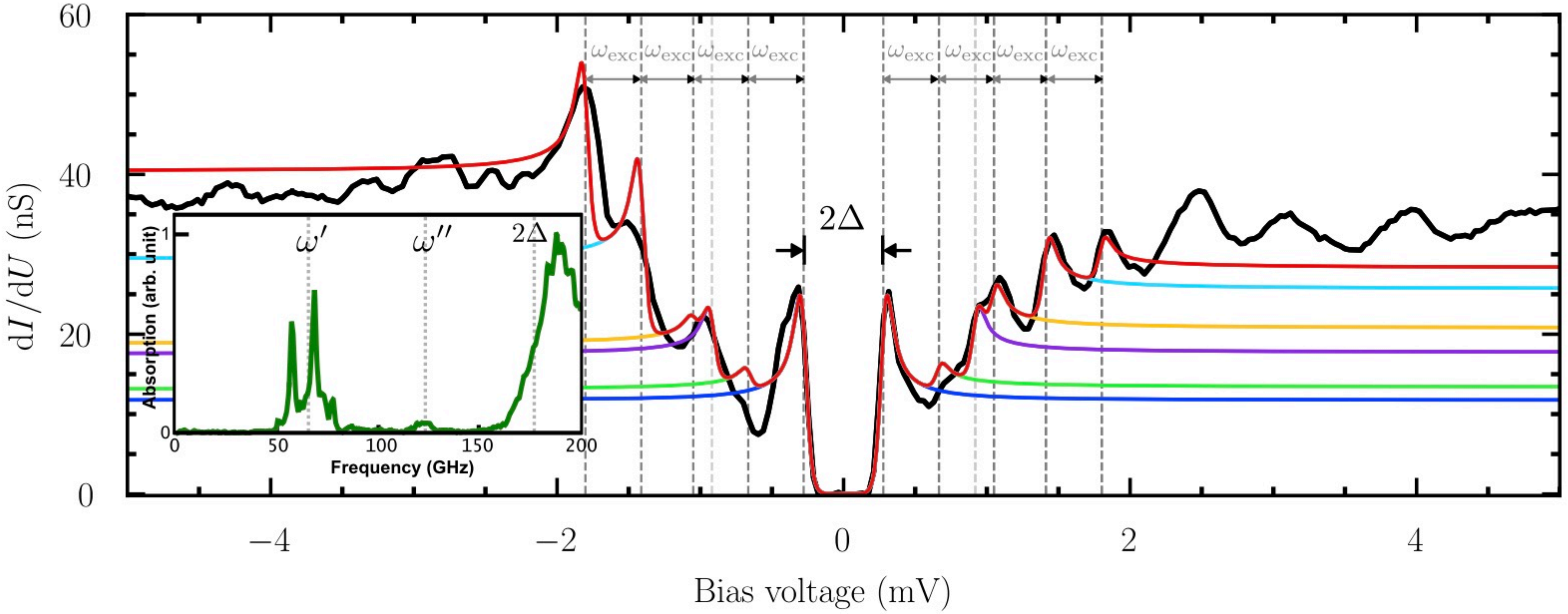

Contributions

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STM experiments & semi-classical calculations


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grAl growth


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DFG Deutsche Forschungsgemeinschaft

Alexander von Humboldt
Stiftung / Foundation

Microscopic view on grAl

- STM transport measurements with sub grain lateral resolution
- Test microscopic theory

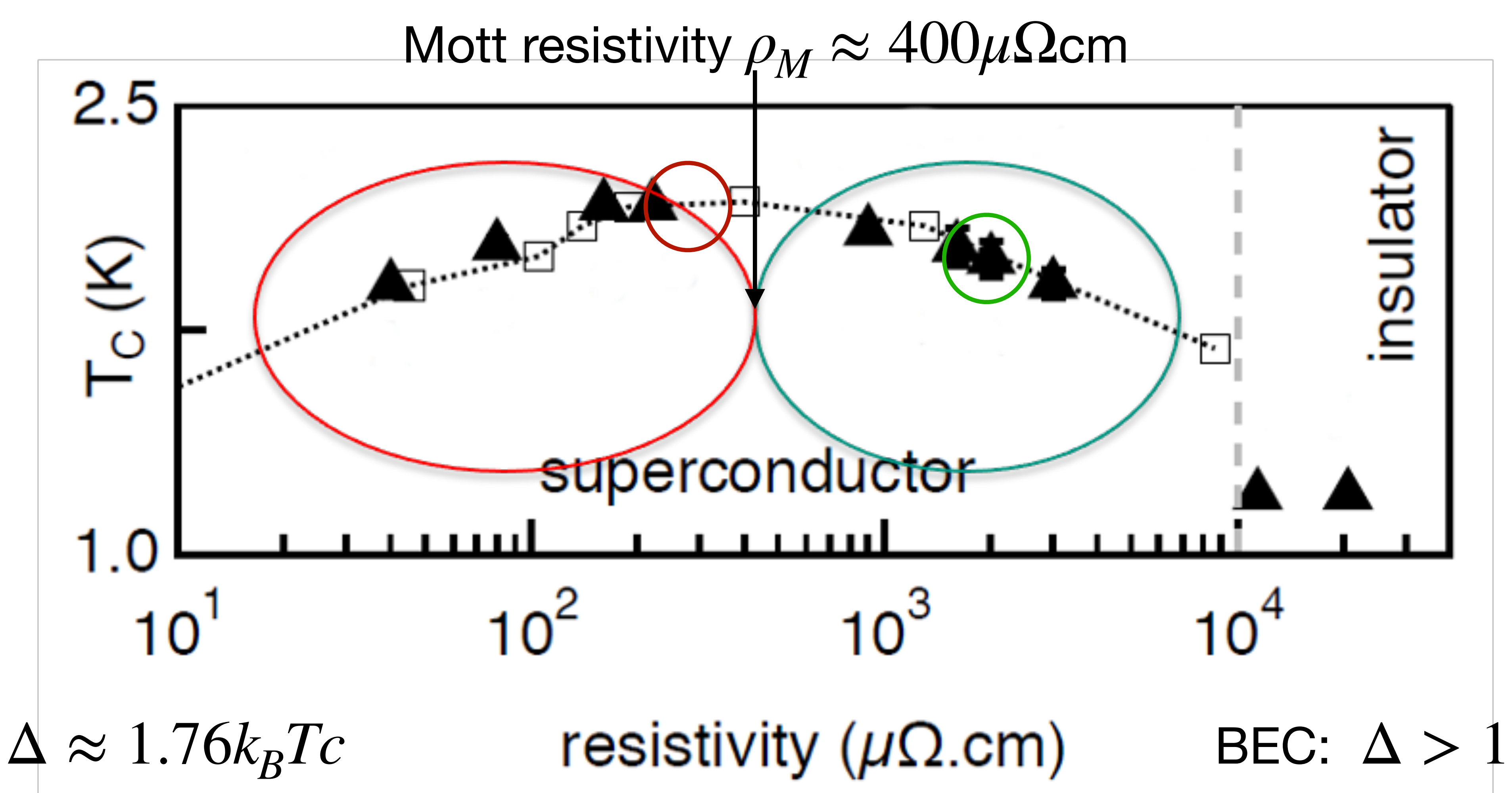
- Superconducting gap and inhomogeneities
- Charging energies

- In gap states and decoherence

- Transfer of samples from growth camber to UHV STM under UHV conditions

pure Al → oxygen poor (metallic) samples → oxygen rich samples

Mott resistivity $\rho_M \approx 400\mu\Omega$cm



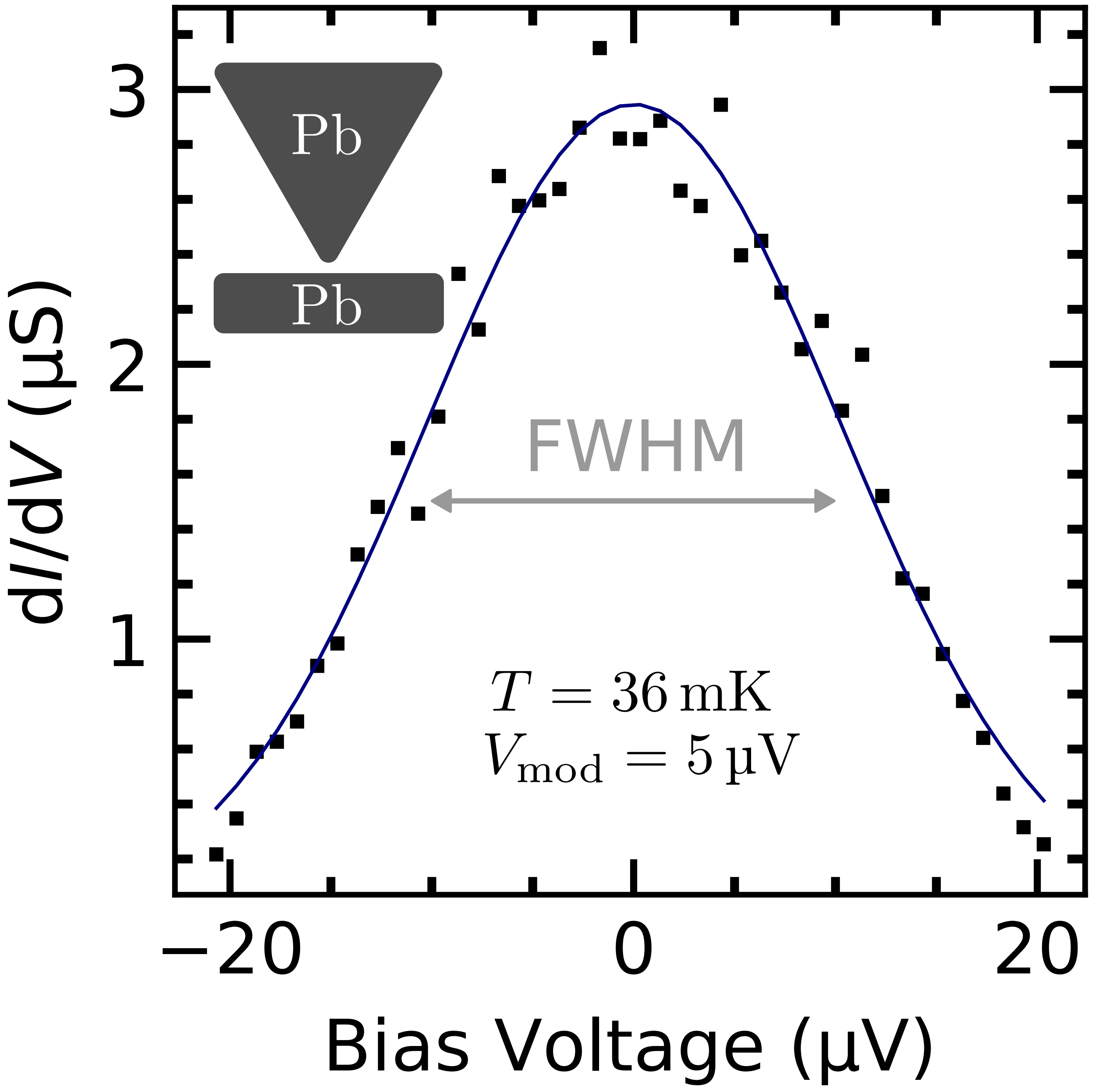
BCS: $\Delta \approx 1.76 k_B Tc$
Cooper pair breaking

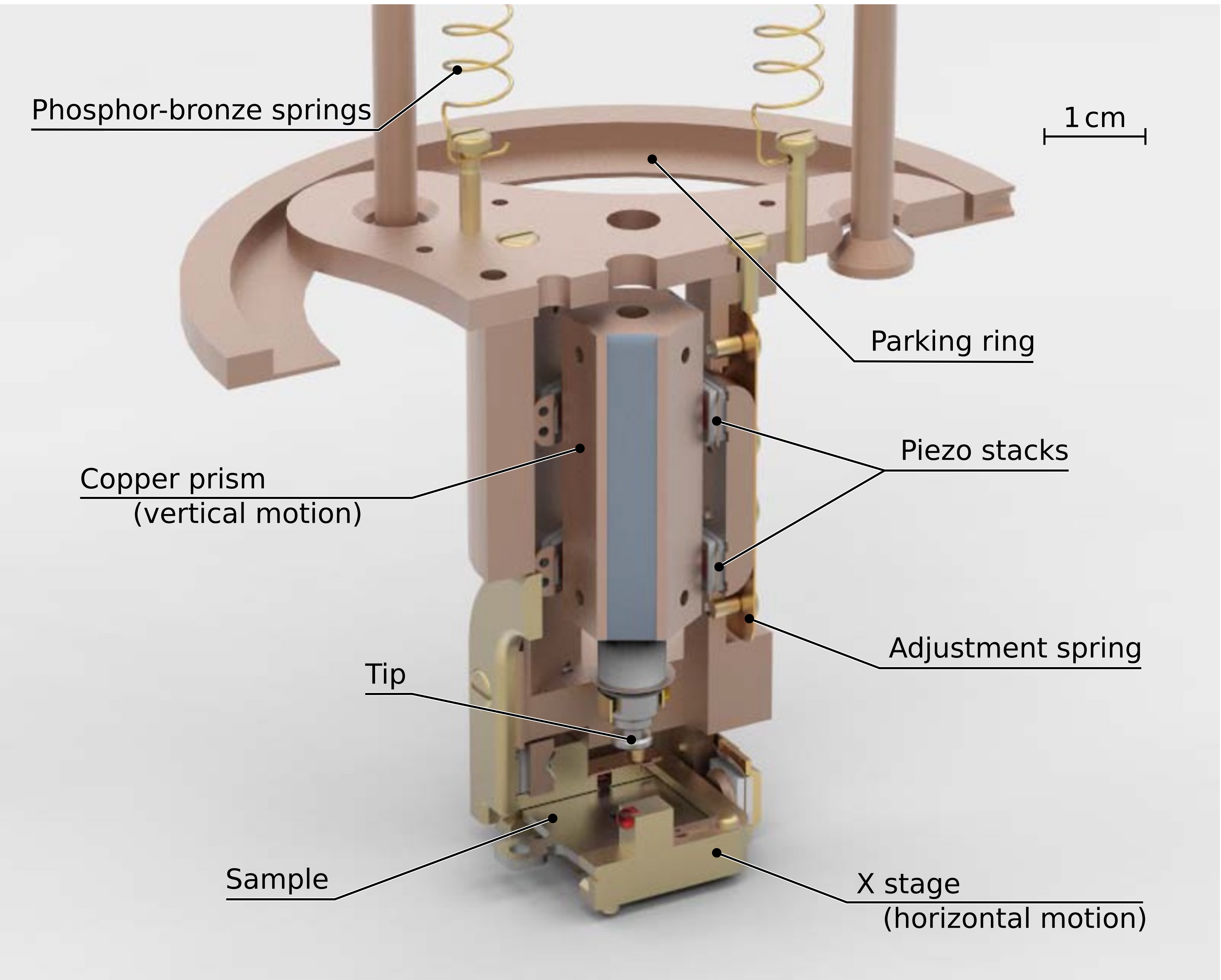
BEC: $\Delta > 1.76 k_B Tc$
Phase fluctuations

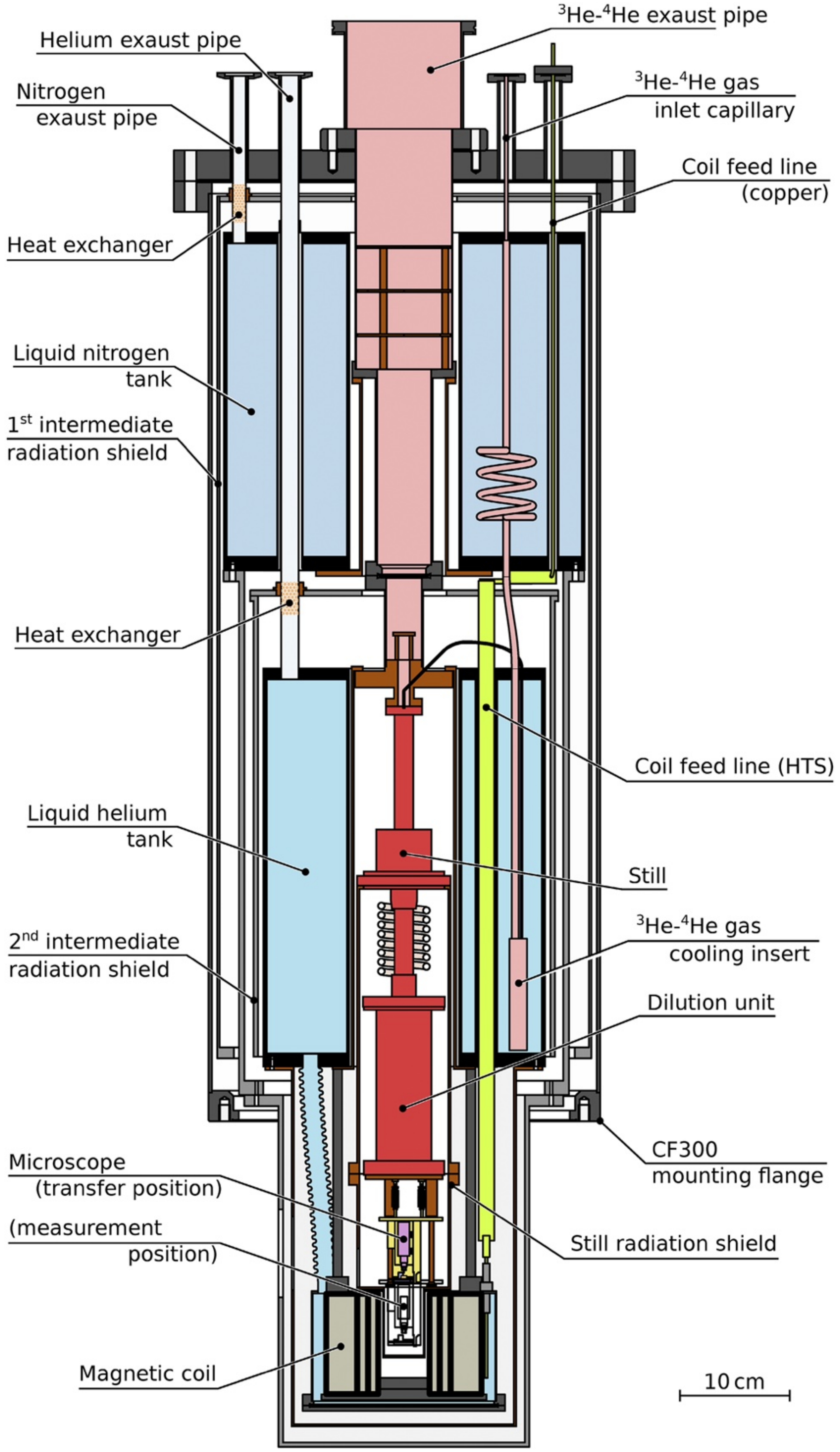
The experimental setup

- Compact and low cost 25 mK STM in UHV
- Magnetic field up to 6.5 T
- 69 mK electronic temperature
- 23 µeV resolution
- <2h sample turn-around

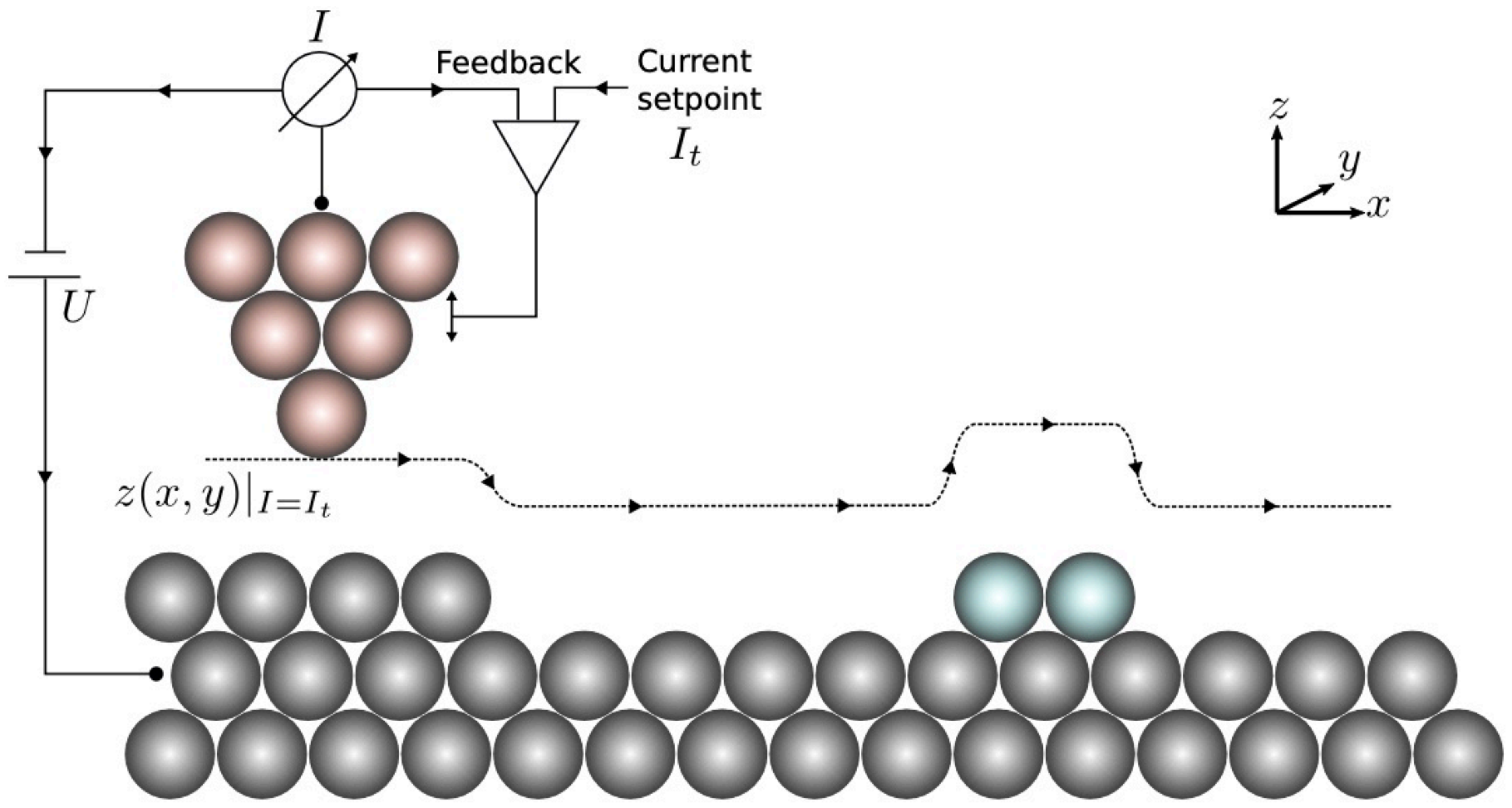
T. Balashov et al. Rev. Sci. Instr. 89, 113703 (2018)

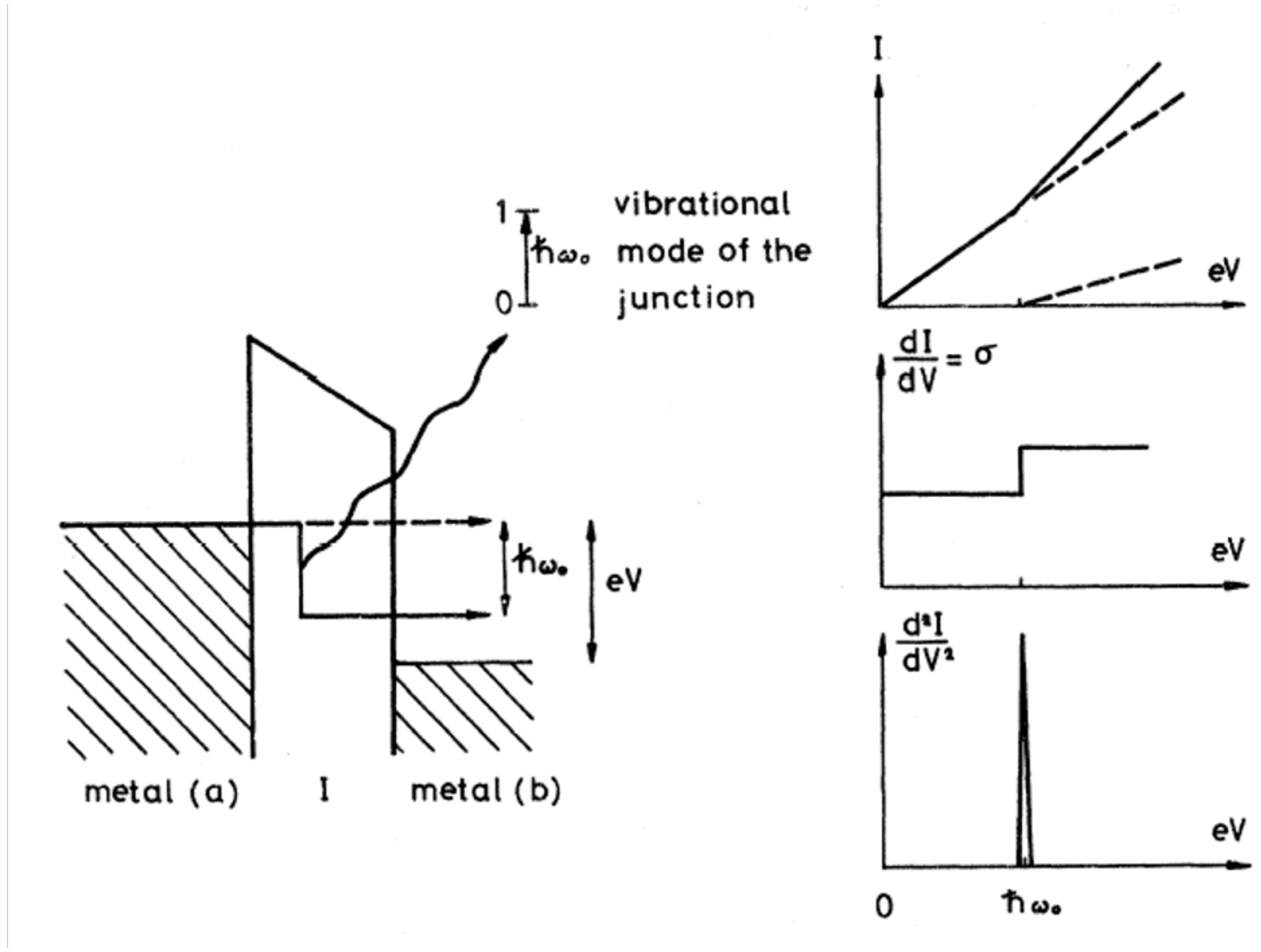






What information do we get from the tunneling current?





- For elastic tunneling with a normal tip, we obtain single particle LDOS

$$\sigma^{\mathrm{el}}(U) = \frac{\mathrm{d}I^{\mathrm{el}}(U)}{\mathrm{d}U} = -\sigma_0 \int_{-\infty}^{\infty} \mathrm{d}\epsilon \tilde{\nu}_{\mathrm{s}}(\epsilon) n_F'(\epsilon - eU)$$

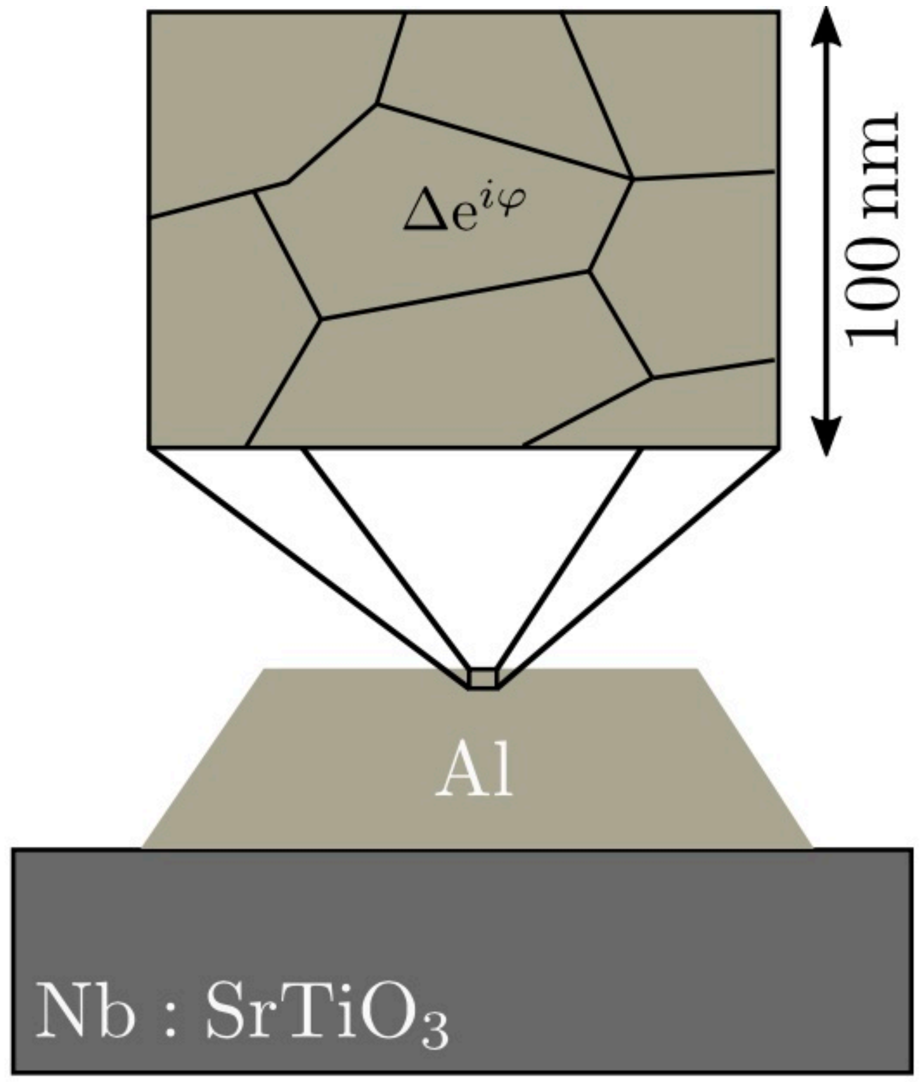
- For inelastic tunneling, final state of electron and excited boson share total energy and momentum of tunneling electron

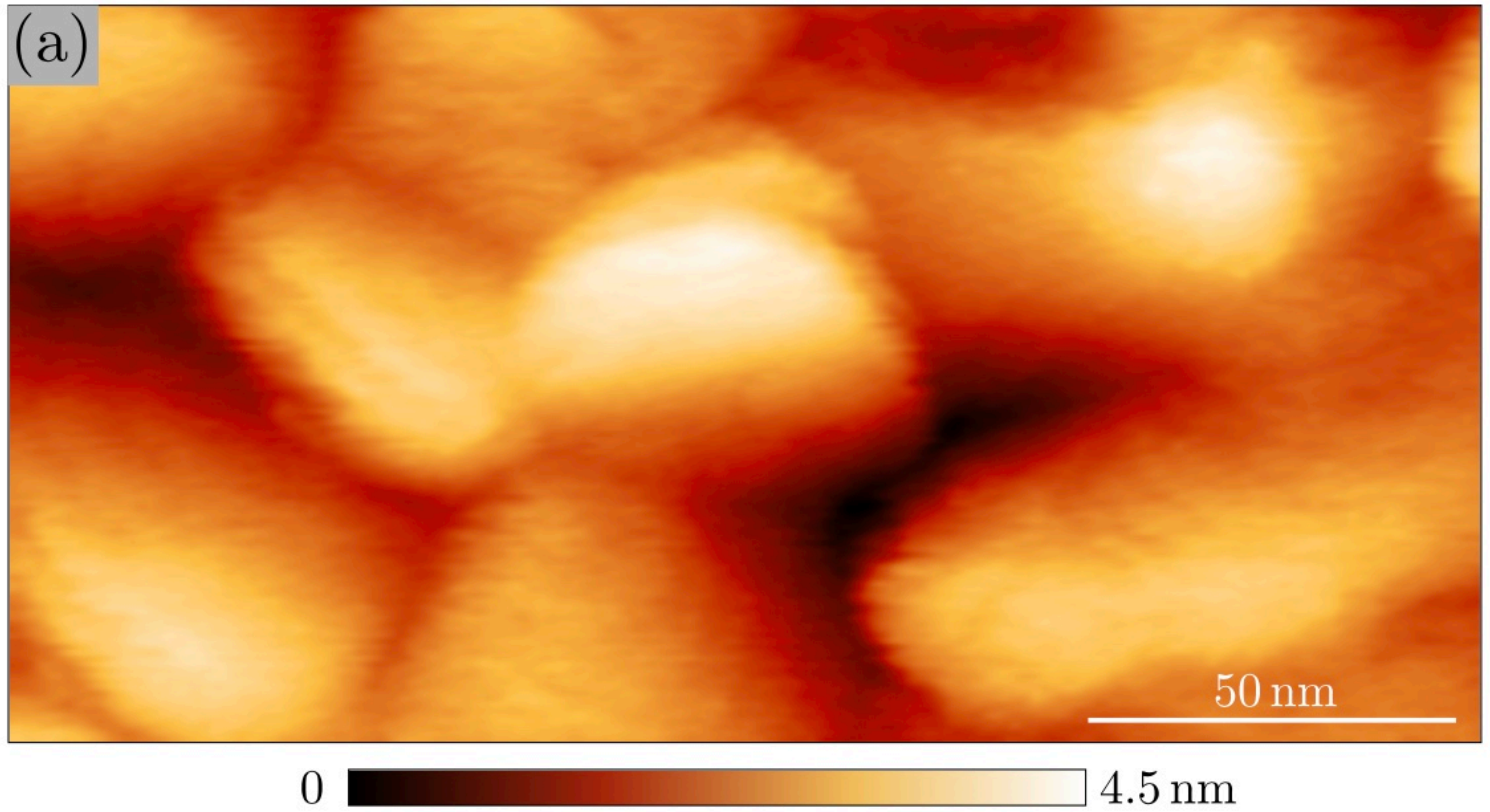
$$\sigma^{\mathrm{inel}}(U > 0) \overset{T \to 0}{\to} \frac{\sigma_0}{\nu_{\mathrm{s}}^F D^2} \int_0^{eU} \mathrm{d}\omega \alpha^2 F(\omega) \tilde{\nu}_{\mathrm{s}}(eU - \omega)$$

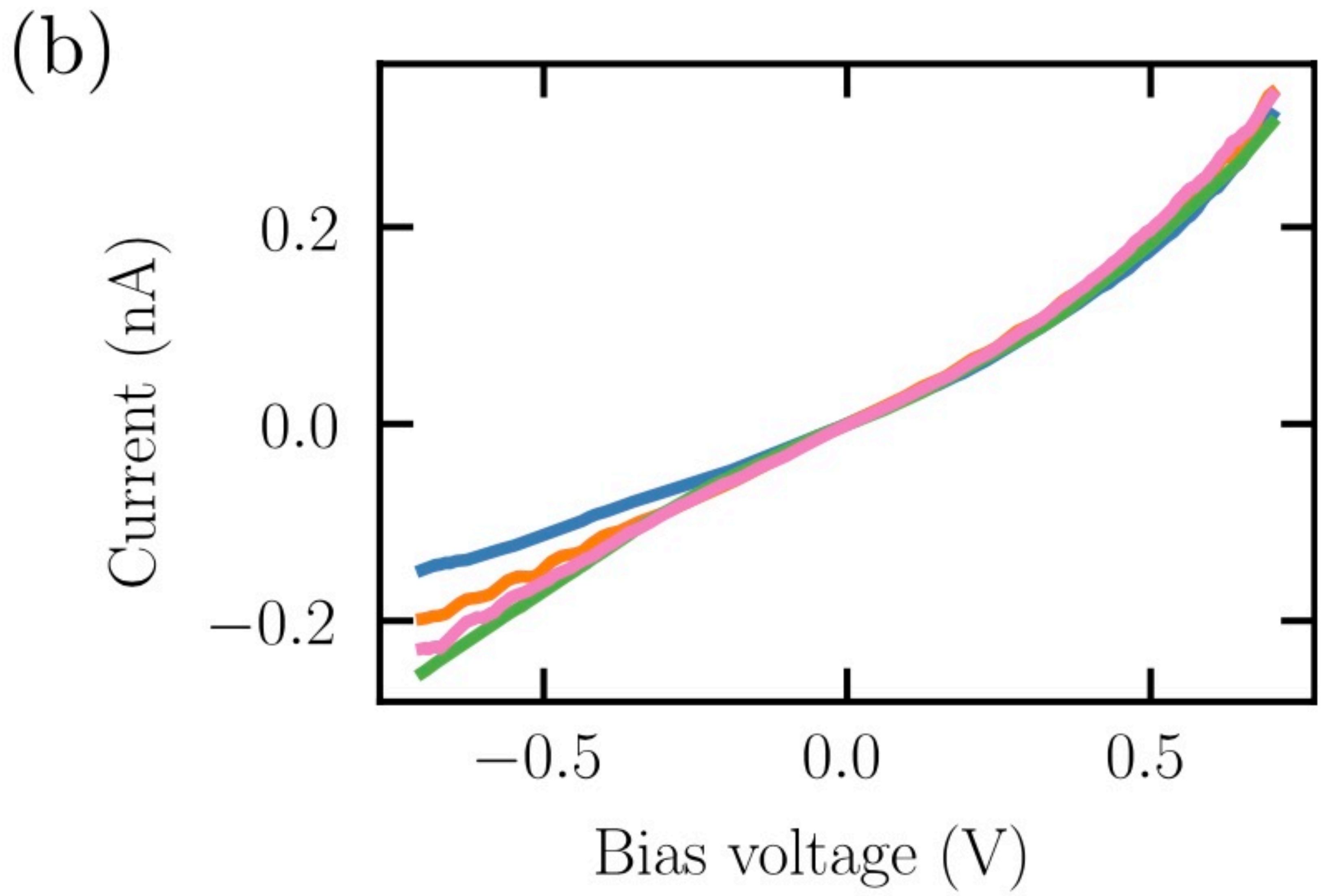
- When sample is superconducting, we see excitations at a bias of $eV = \Delta + \omega$

- Full transport should consider addition/subtraction spectrum and path of current Landauer Büttiker

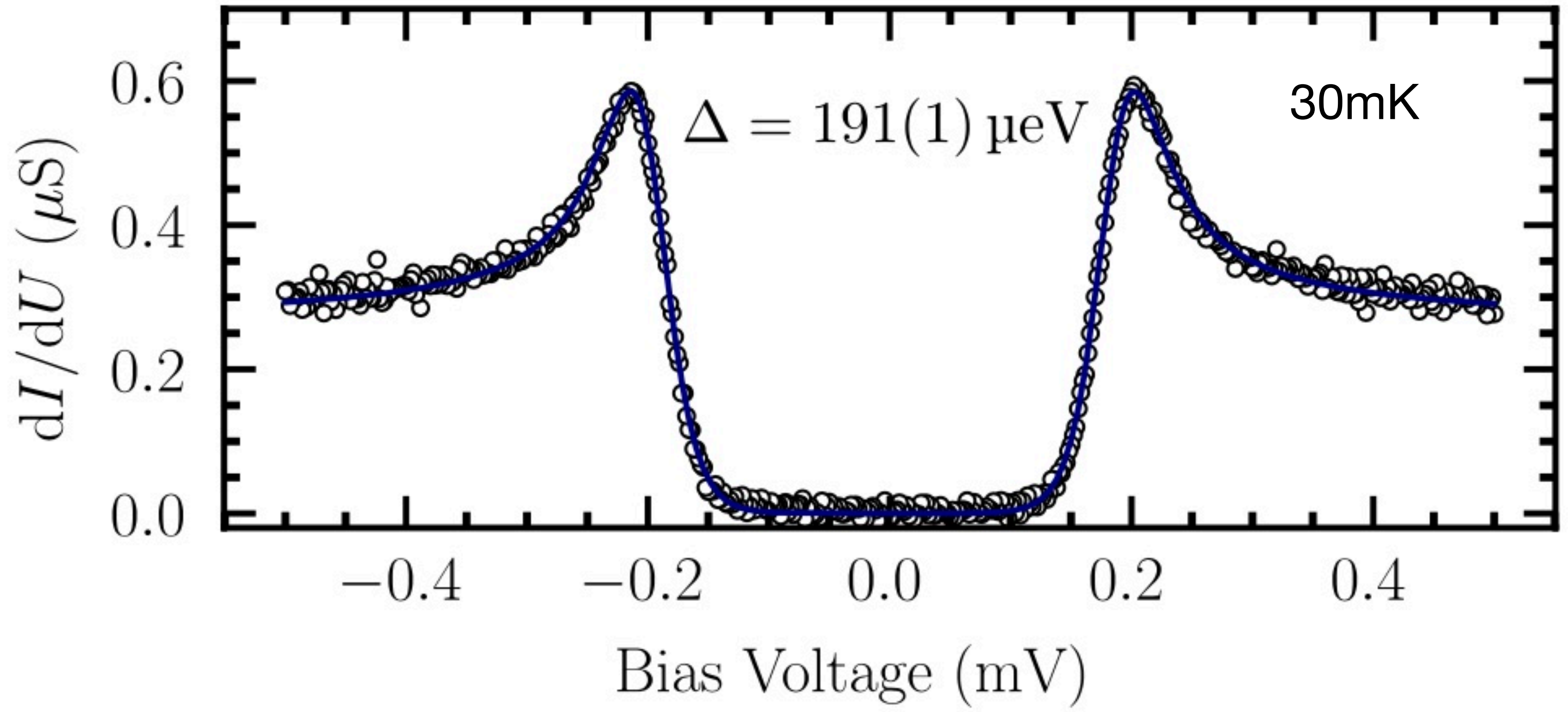
Pure Al



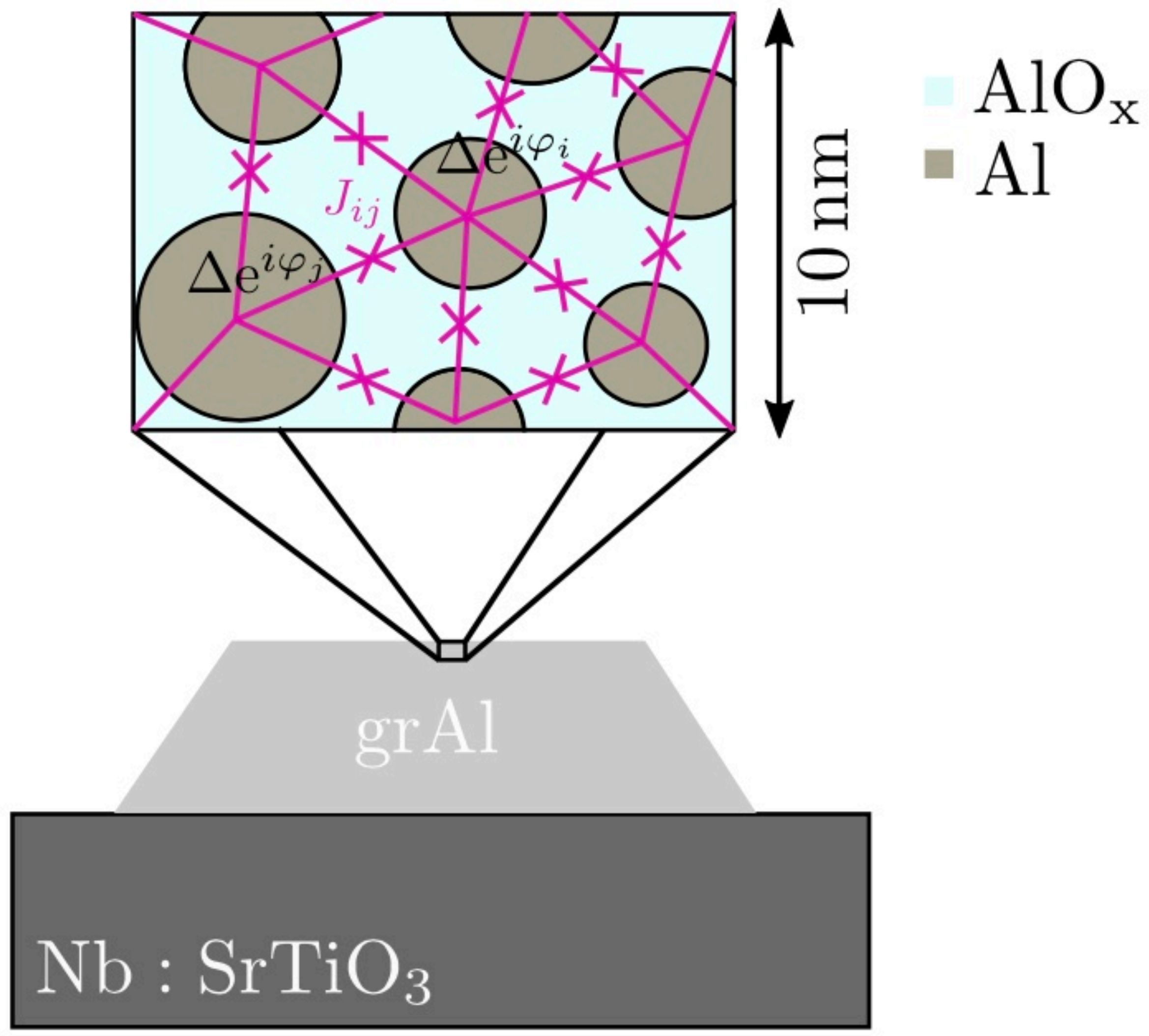


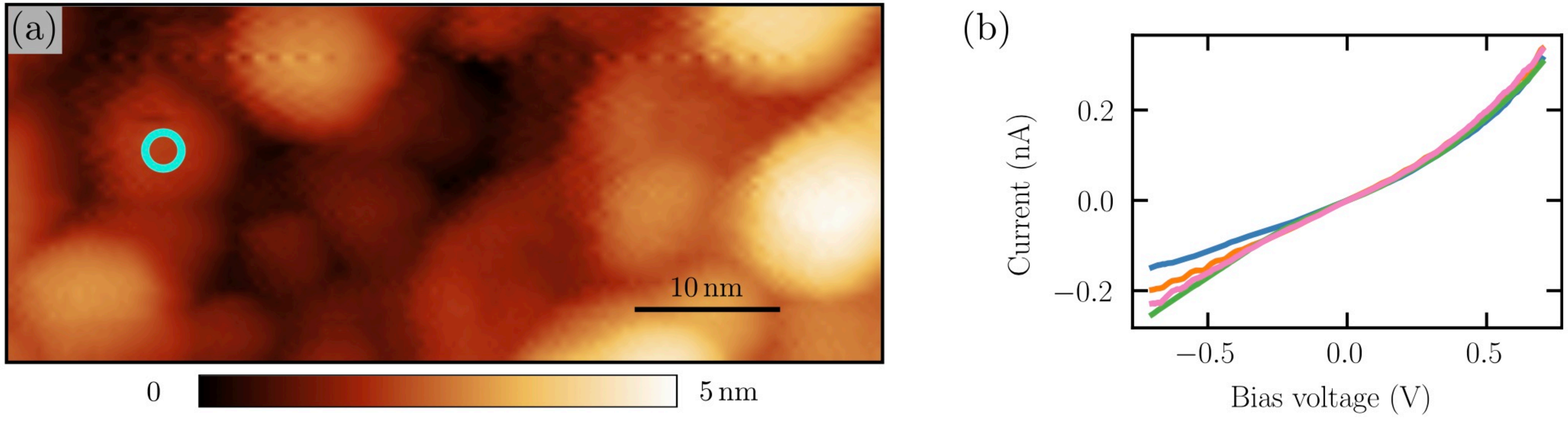


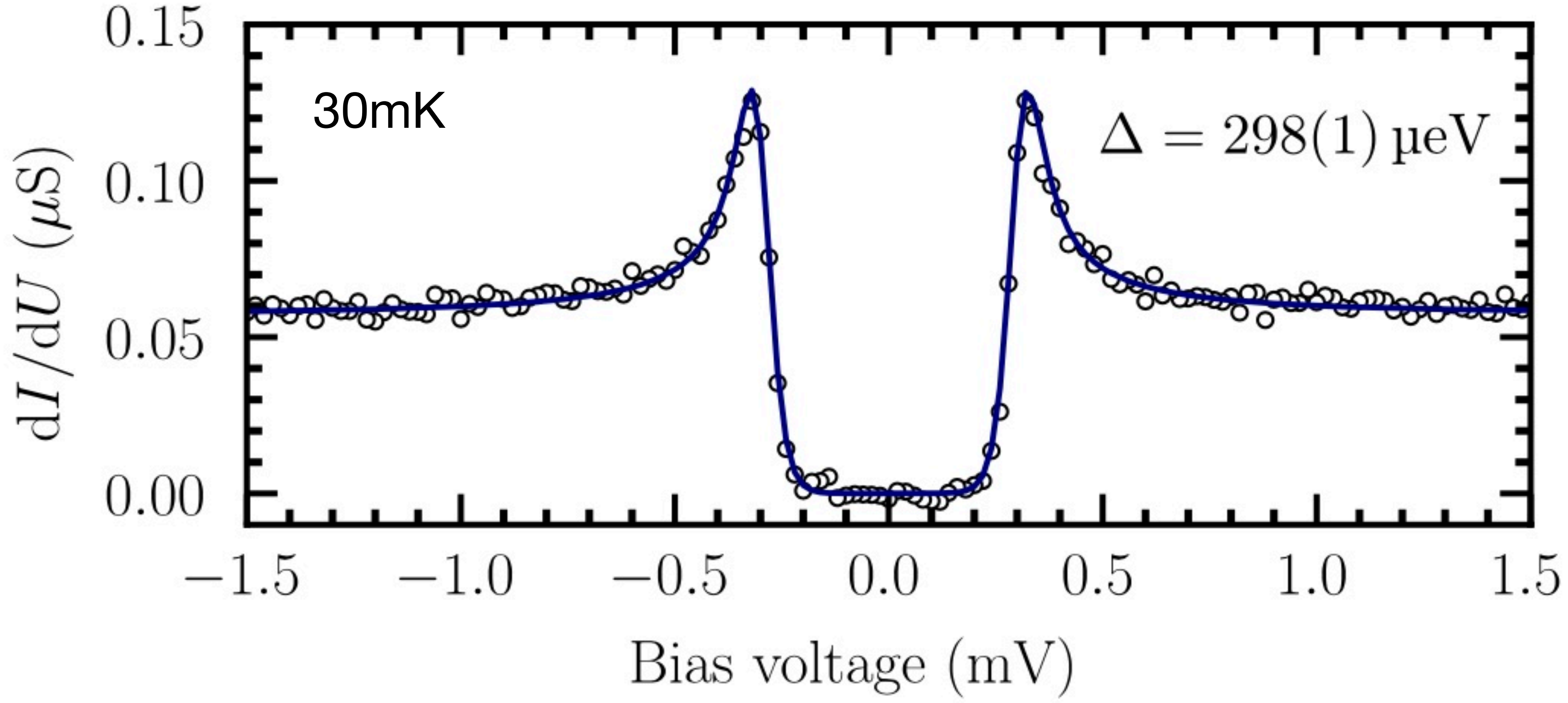
- Efficient Al diffusion leads to large grains
- Metallic I(V) curve
- Gap is close to literature value of Al films (200μeV)
- Gap is homogeneous and no in-gap states found
- Surface is clean



Oxygen poor samples (300 $\mu\Omega$cm)



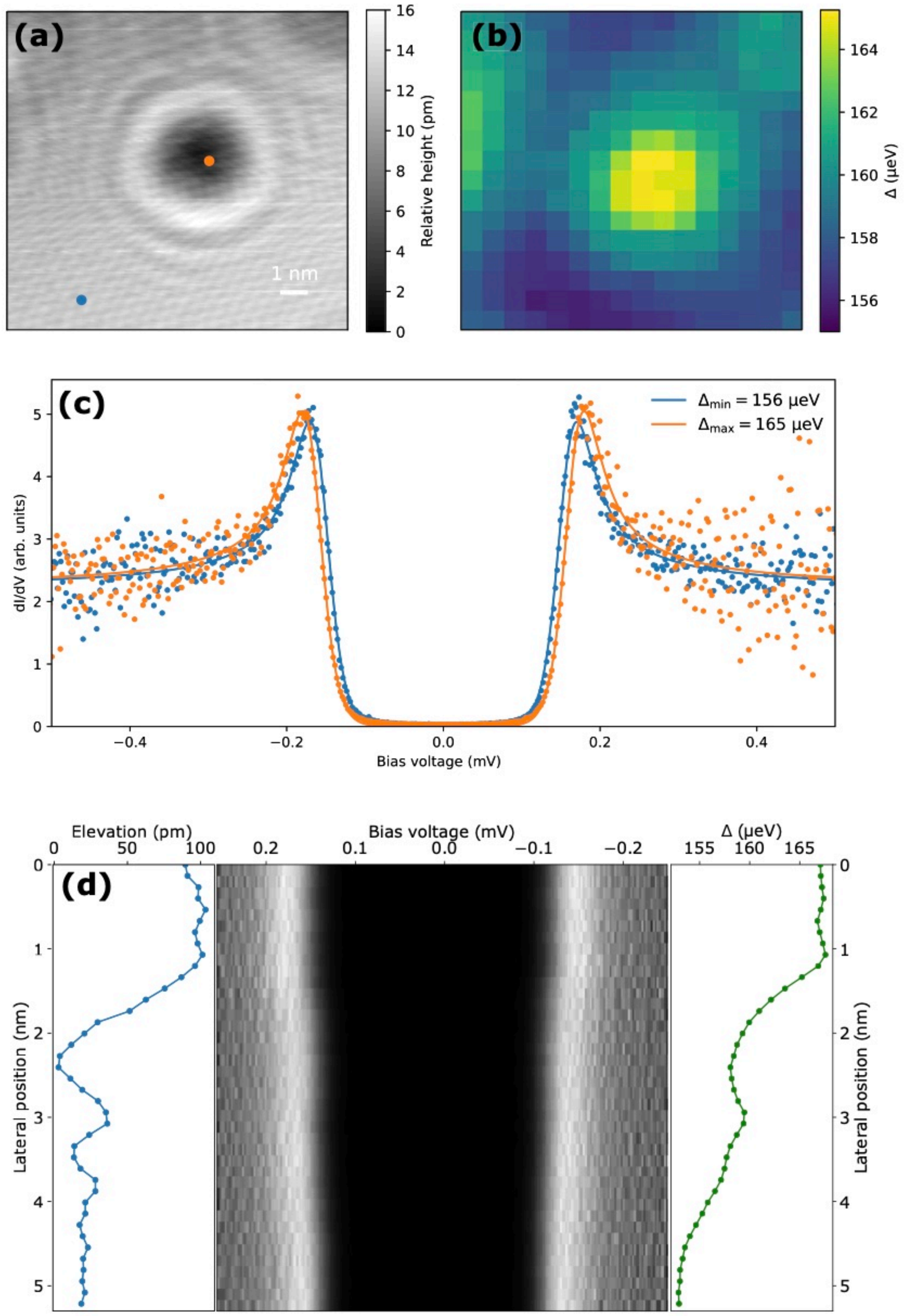




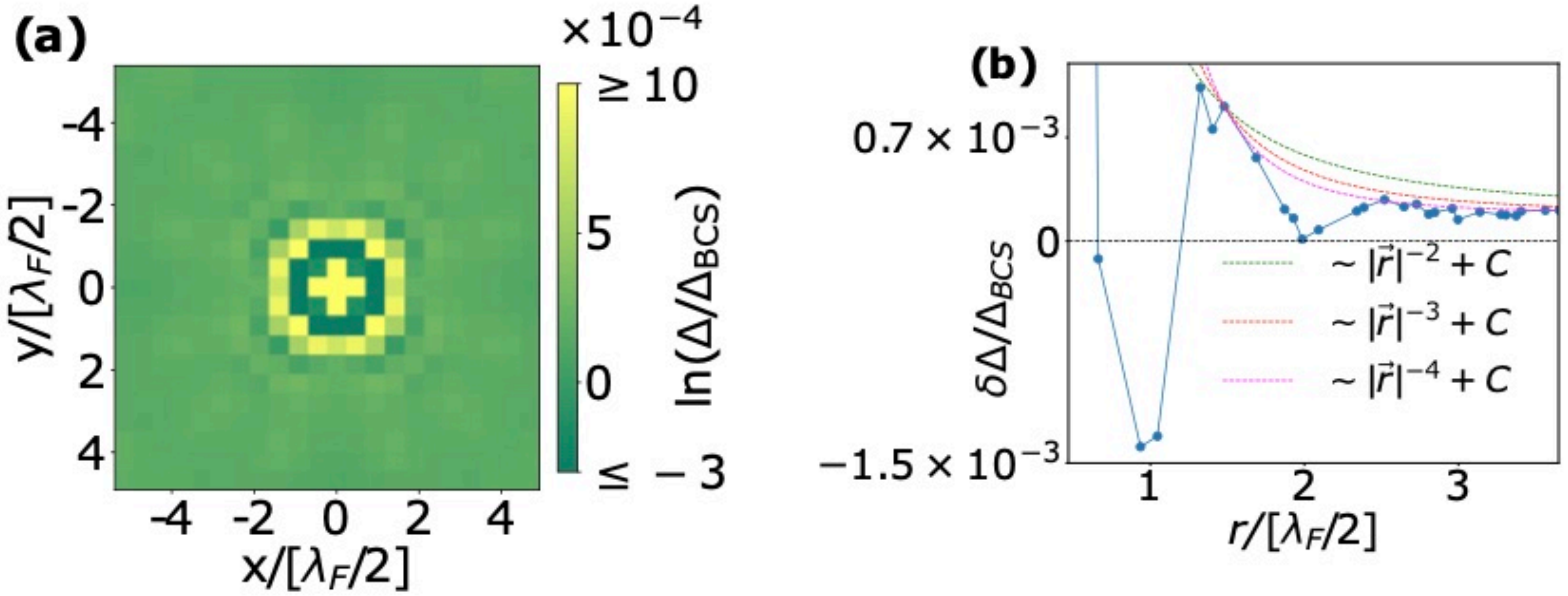
- Al diffusion hampered by AlOx with grains 5-10 nm
- Metallic I(V) curve
- Gap is much larger than pure Al
- Gap is homogeneous and no in-gap states found

Increase of the gap by disorder

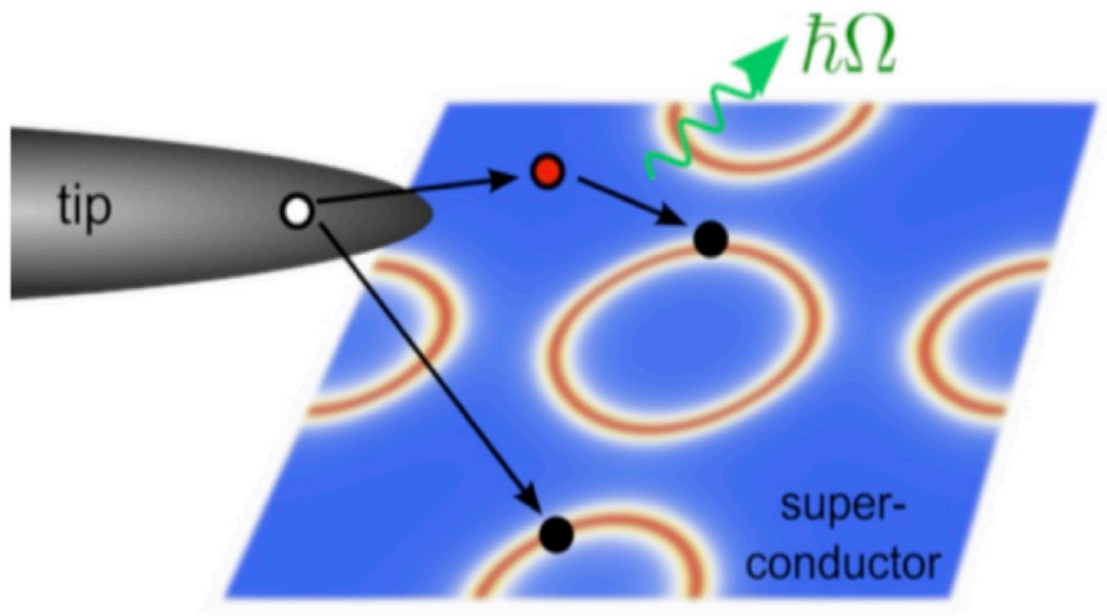
- Single implanted Ar atom in Al(111)
- Local increase of gap by 7%
- Damped Friedel oscillations in gap
- Reproduced in model calculations

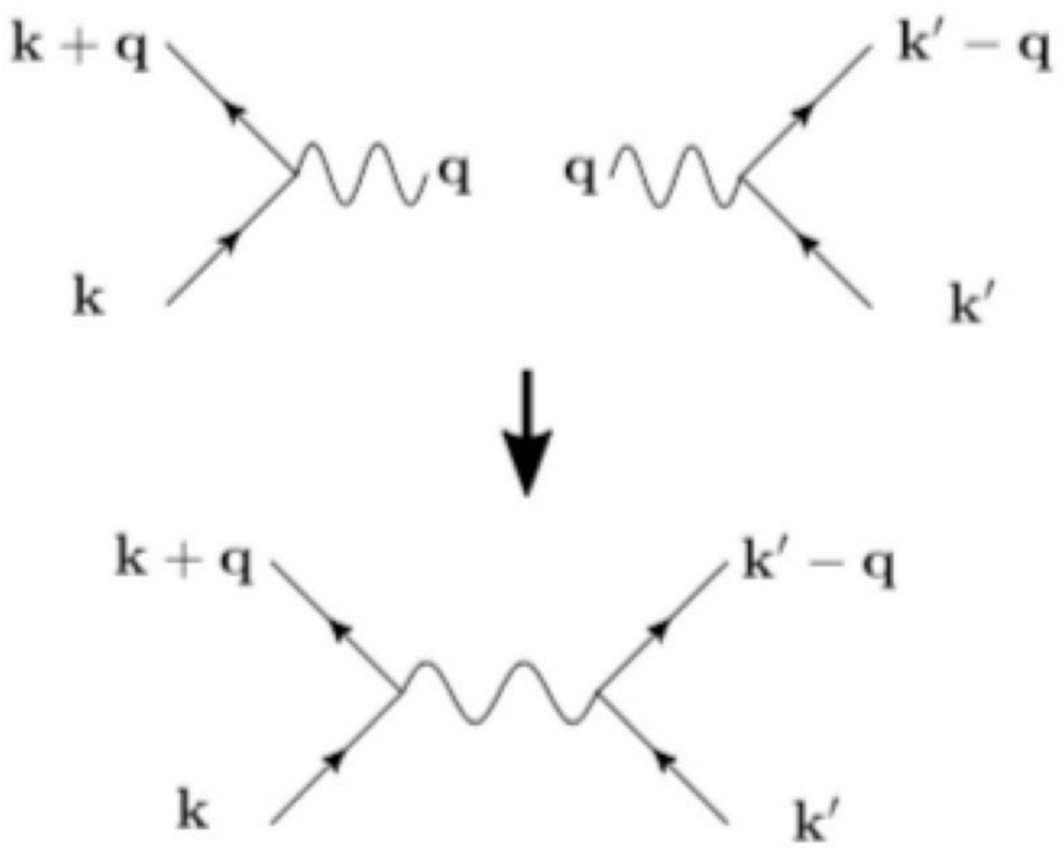


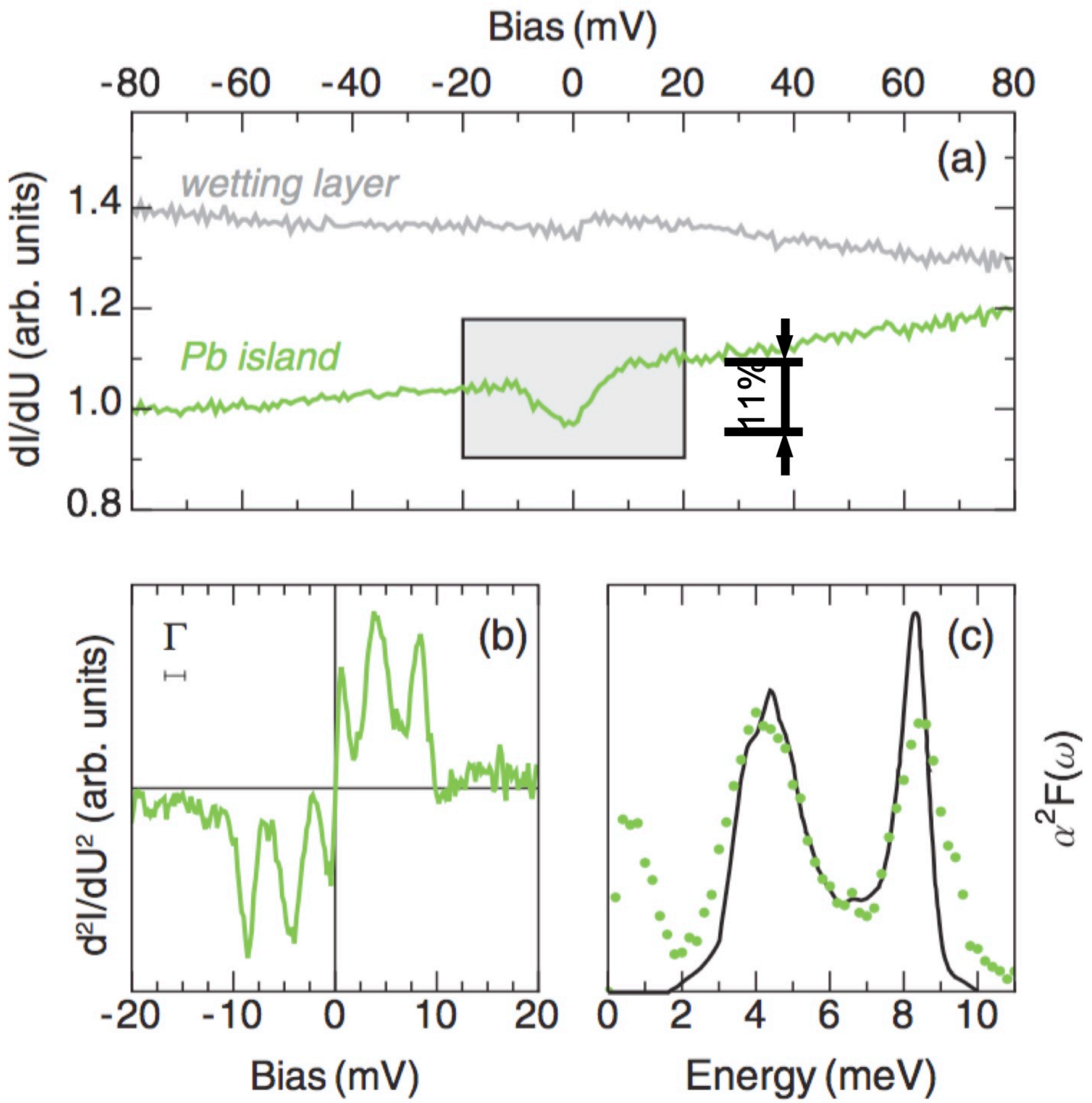
Stosiek et al. PRB 105, L140504



Detecting phonons with ITS



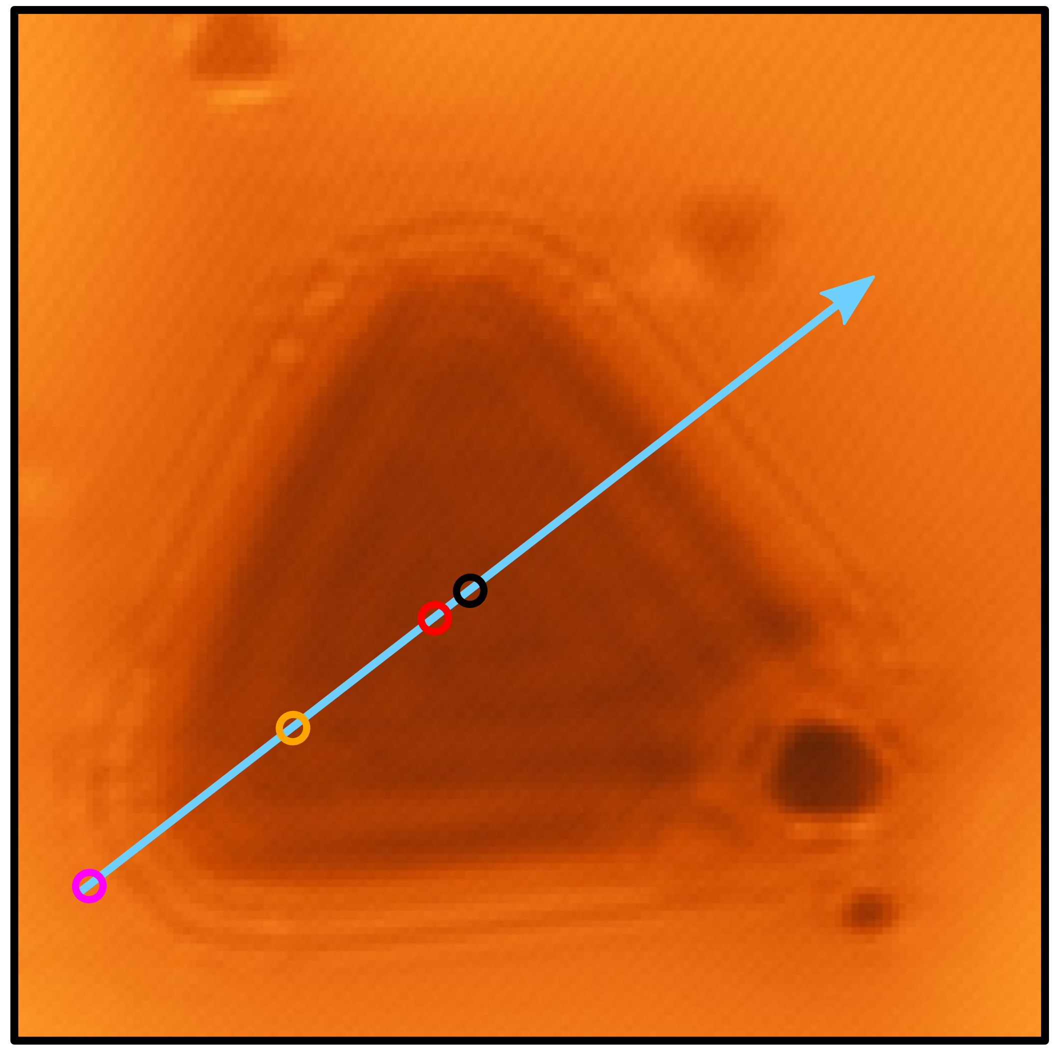


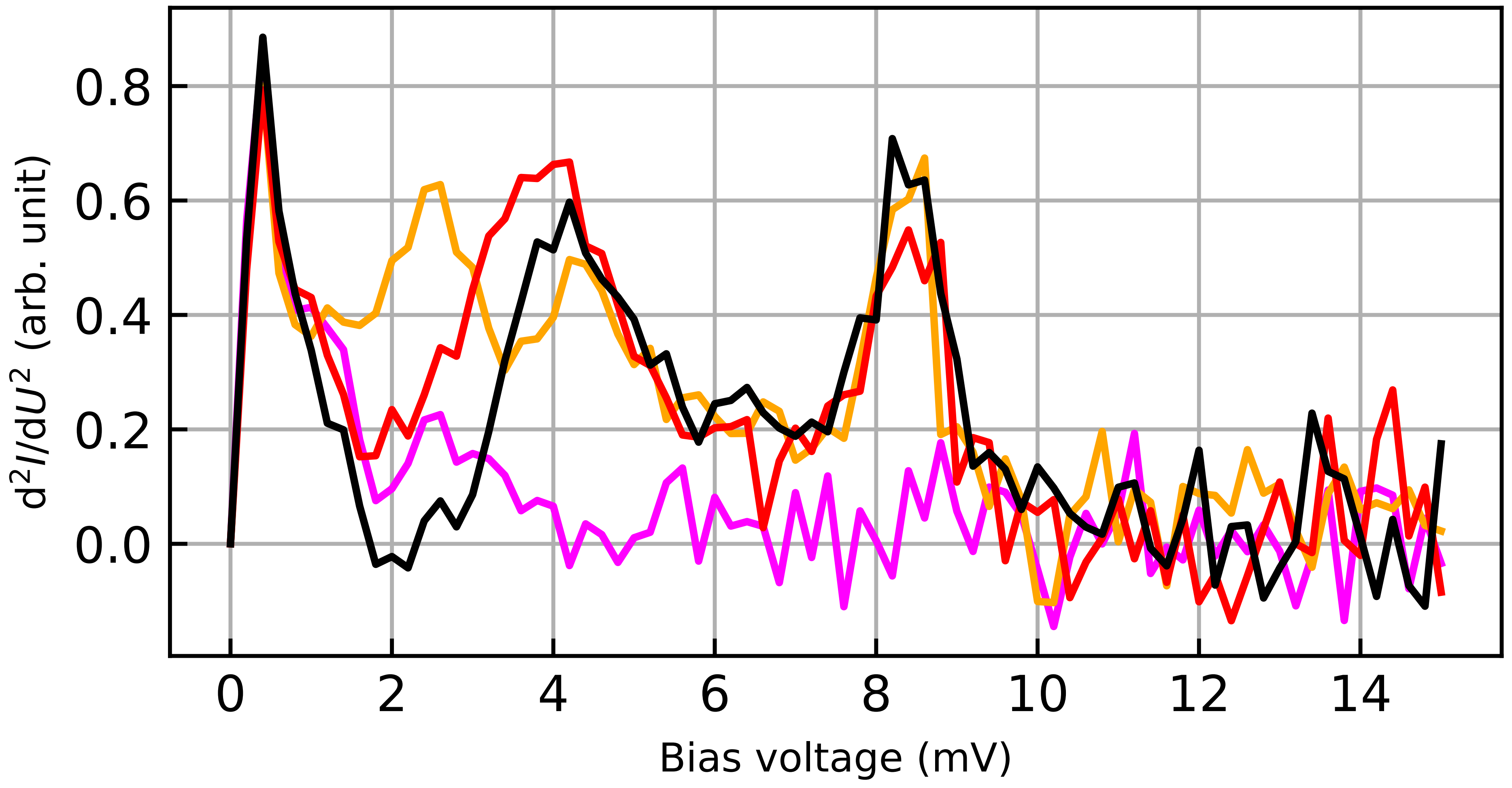


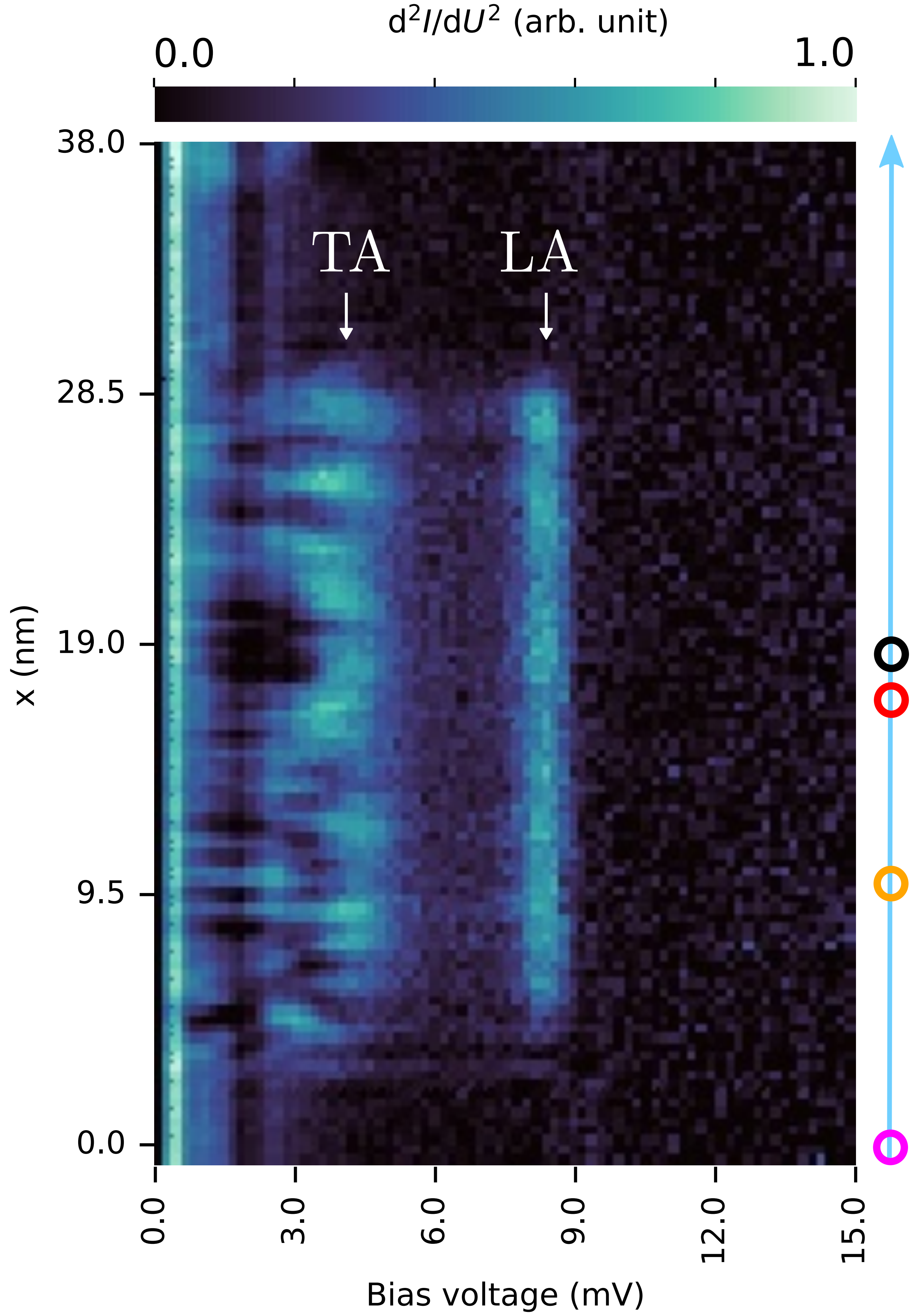
M. Schackert et al., PRL 114, 42002 & J. Jandke et al., PRB R060505

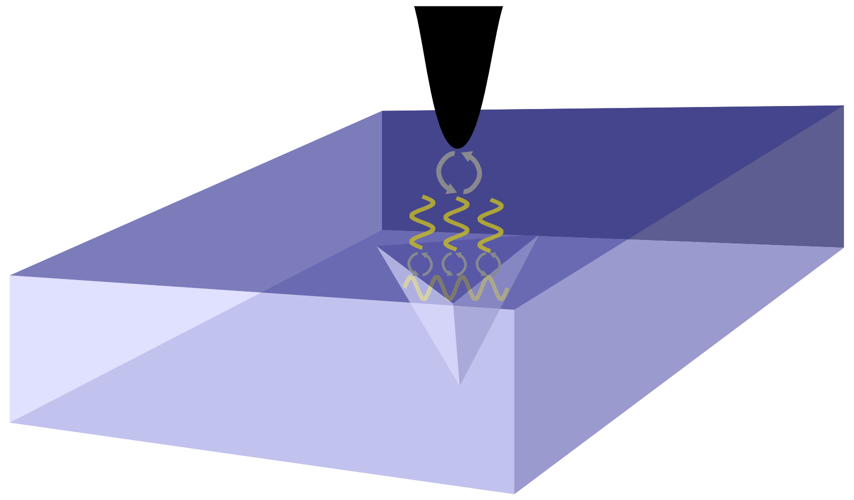
- Inelastic tunneling spectroscopy (ITS): $\dfrac{\partial^2 I}{\partial U^2} \propto \alpha^2 F(\omega)$
- Initial tip state near $k_{\|}^{e} = 0, E$: final on shell sample state: $k_{\|}^{e} = -q_{\|}, E-\omega, q_{\|}^{p}, \omega$
- Phase space for inelastic scattering restricted by momentum conservation
- Can be enlarged by breaking translational symmetry

Stacking fault tetrahedron in Pb(111)



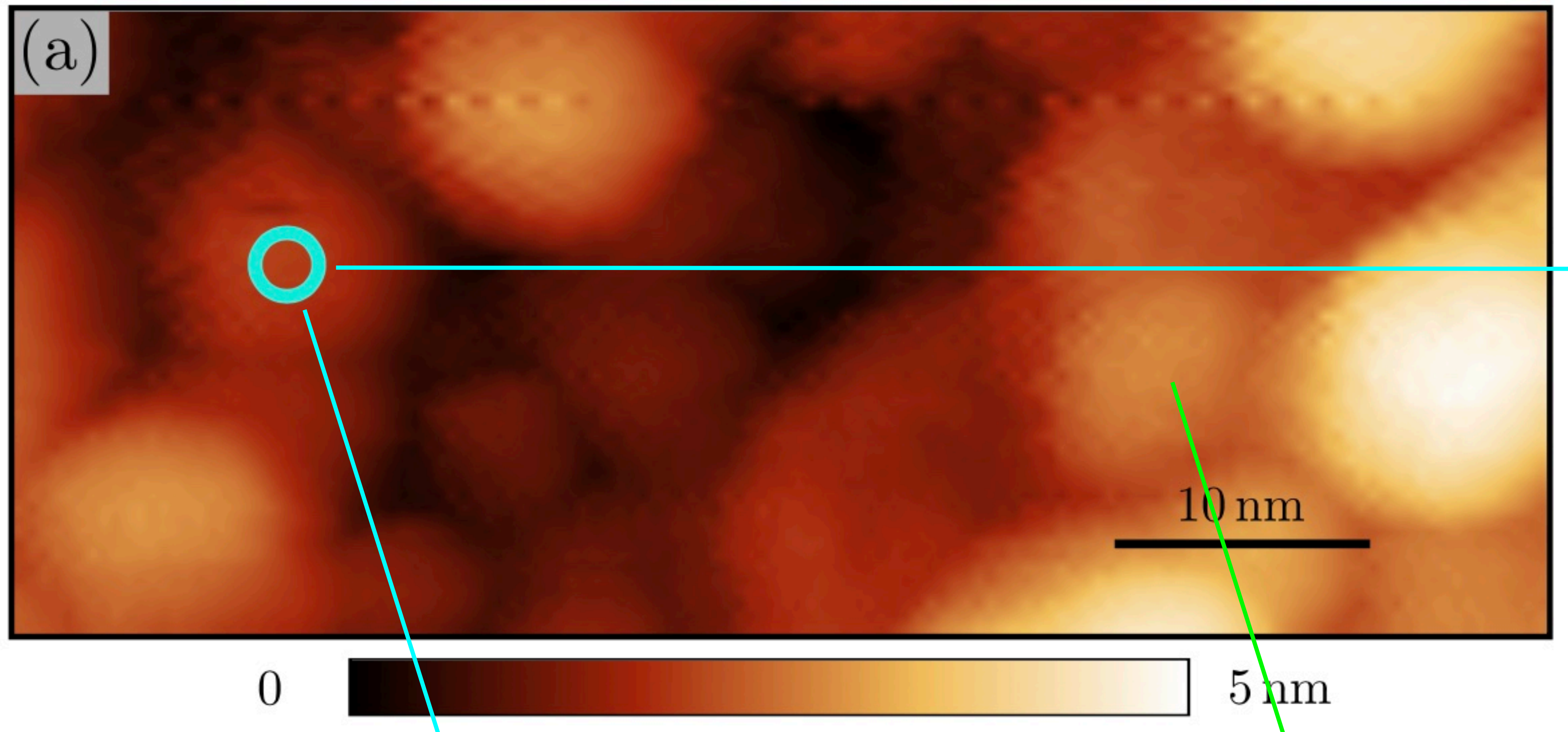


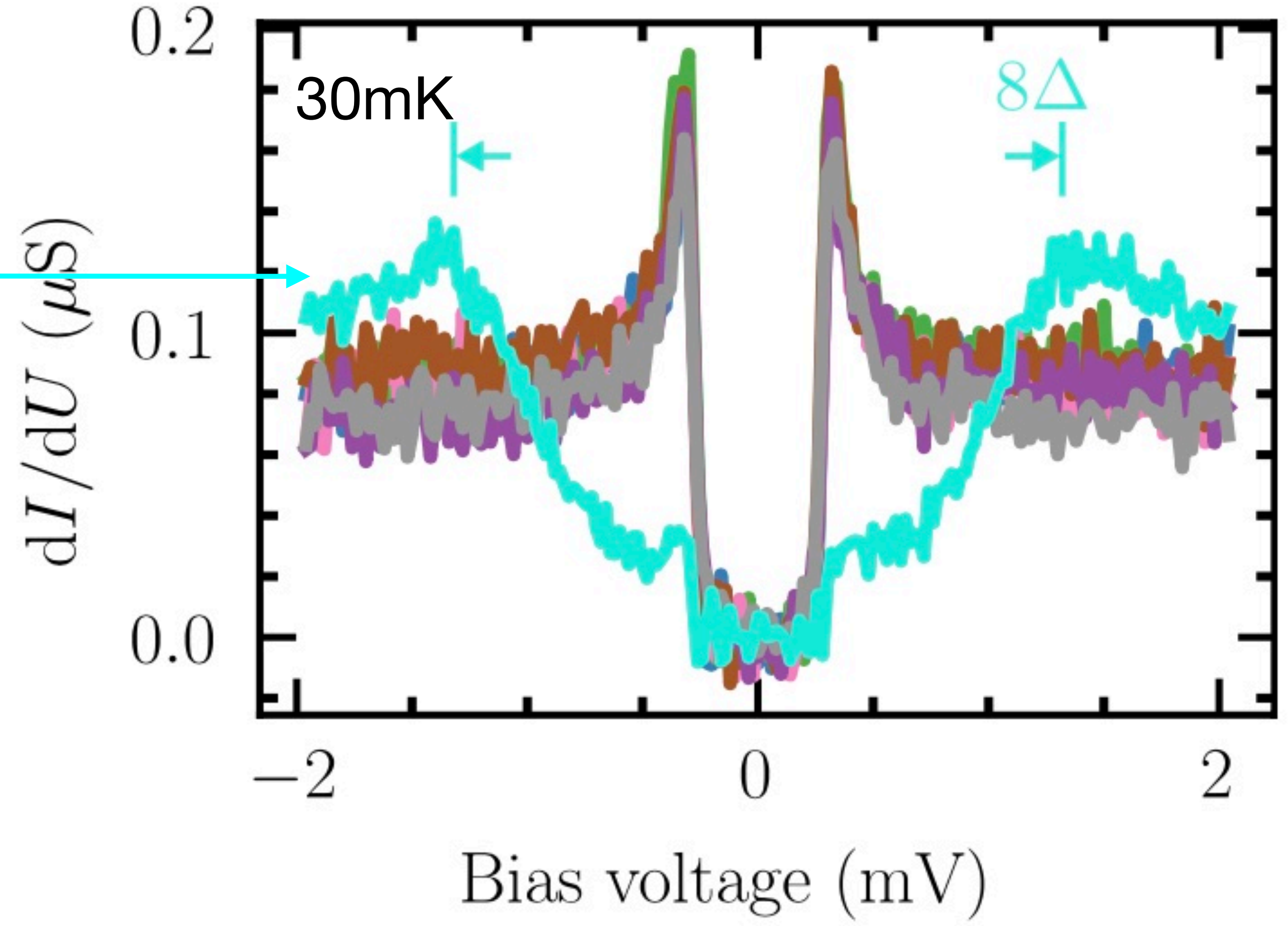


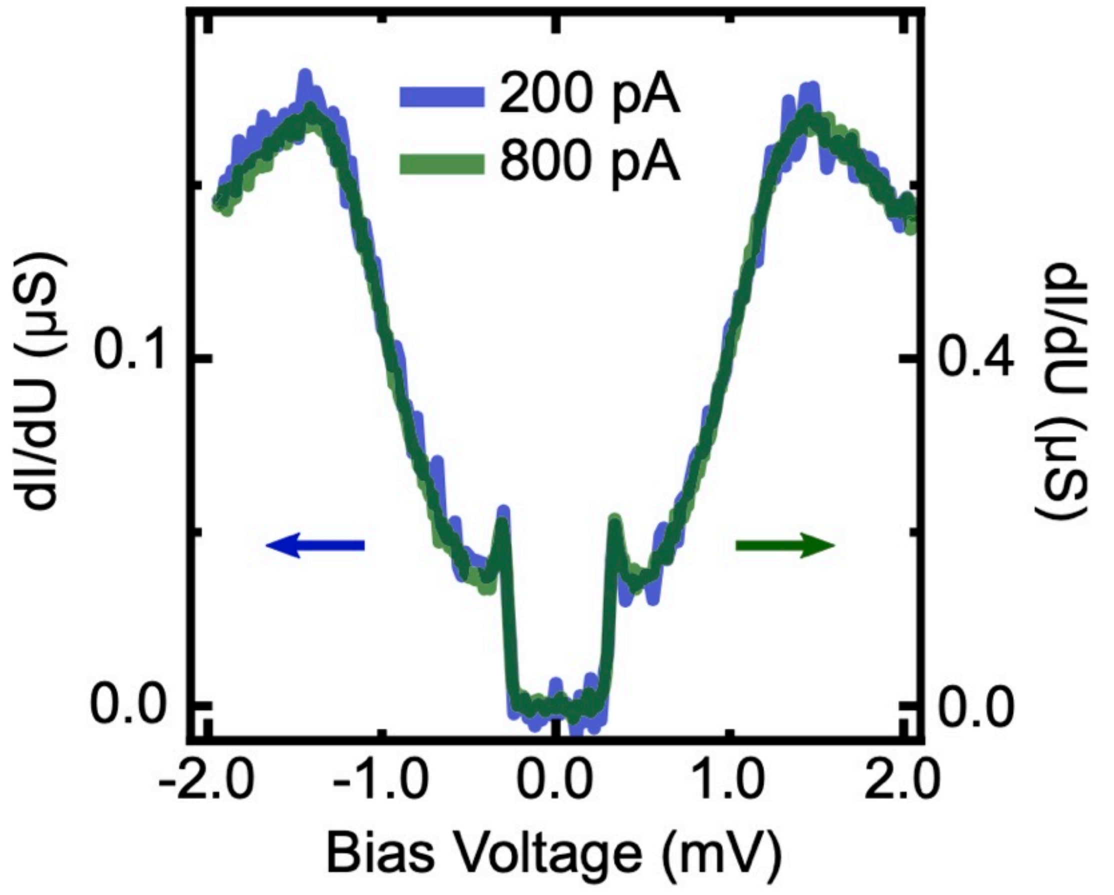


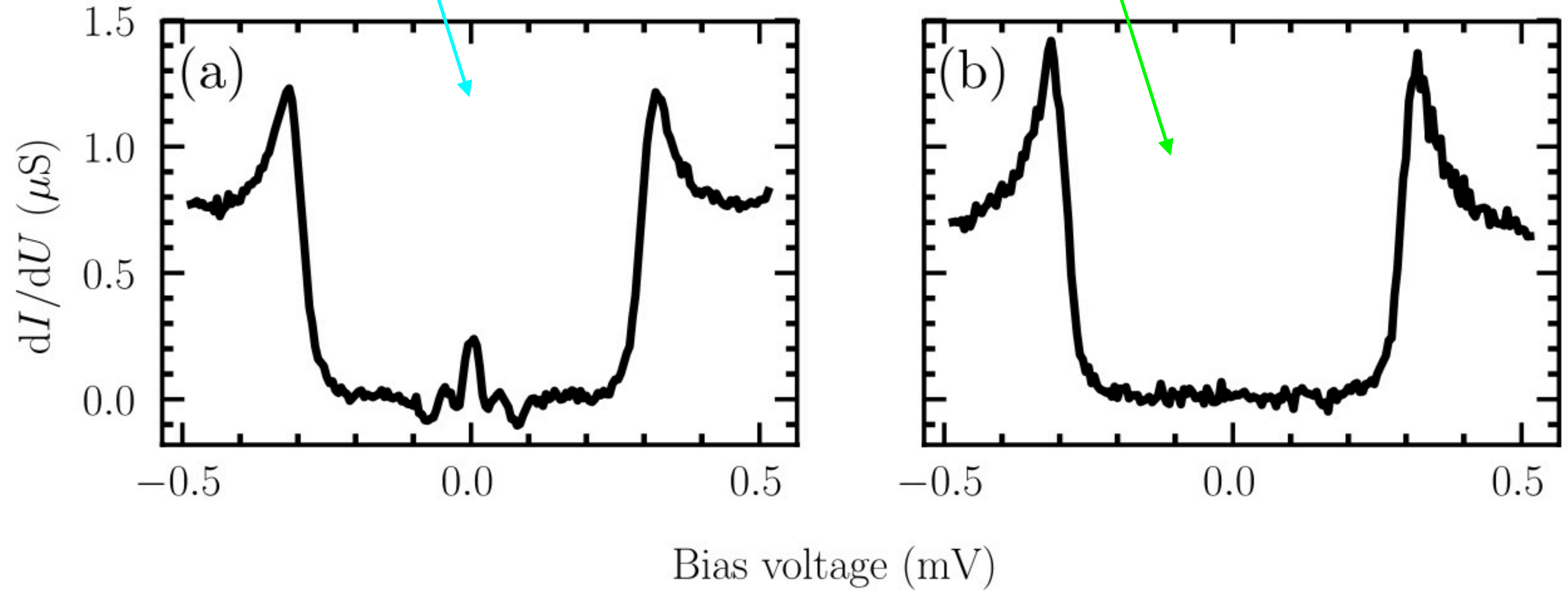
- Enhanced electron-phonon interaction inside tetrahedron
- Clear van Hove peaks for transversal and longitudinal phonon branch
- The tetrahedron acts as a box for standing electrons, lifting the strict selection rules
- Transversal phonons show structure

Oxygen poor samples (300 $\mu\Omega$cm)



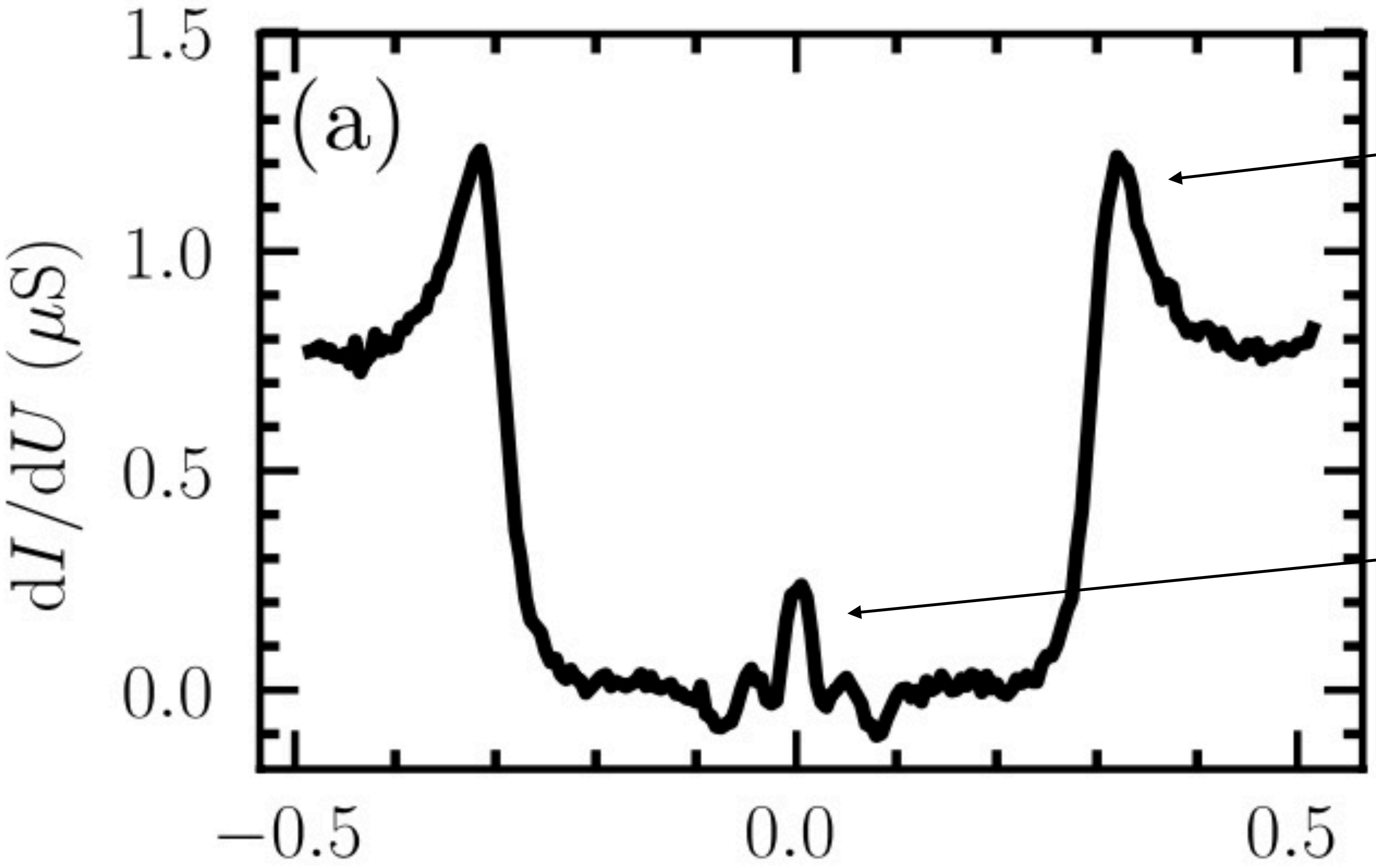






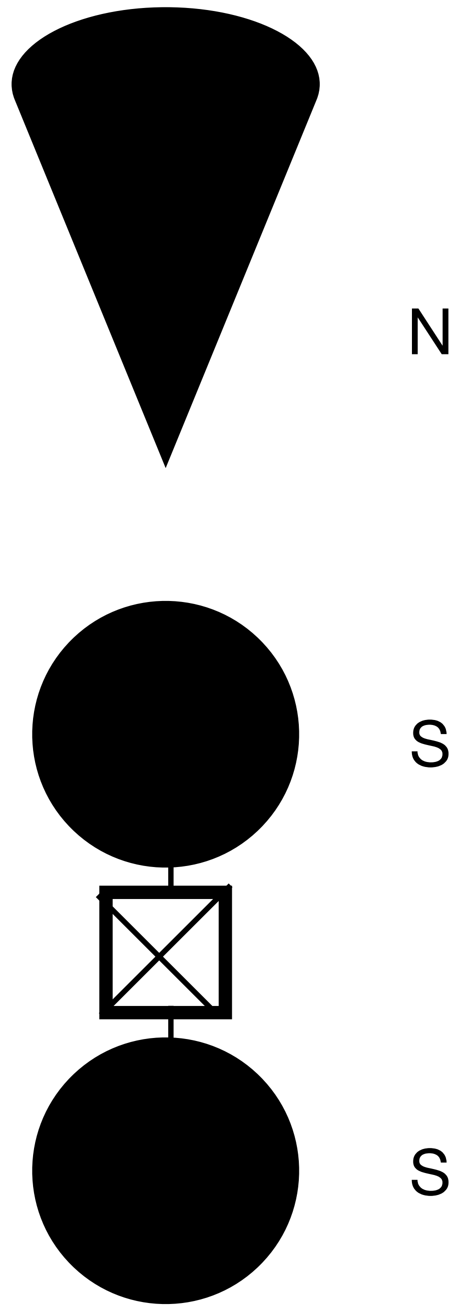
- Few grains show an additional pseudo-gap of ≈8Δ
- Pseudo gap is no function of current
- First signs of decoupling detected at high current densities for grains with pseudo gap
- Josephson coupling seen around 0V

Oxygen poor samples (300 $\mu\Omega$cm)



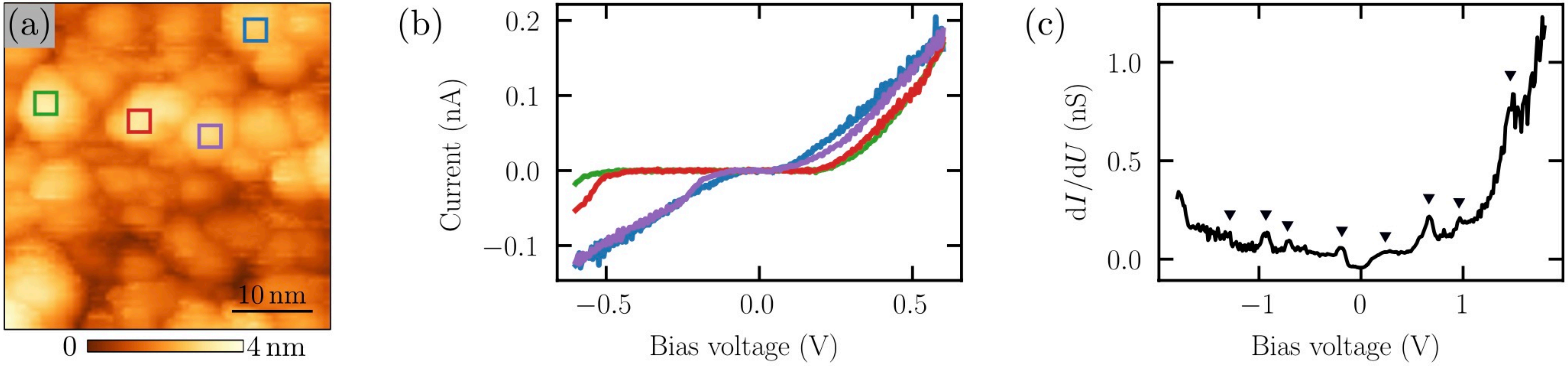
single particle tunneling

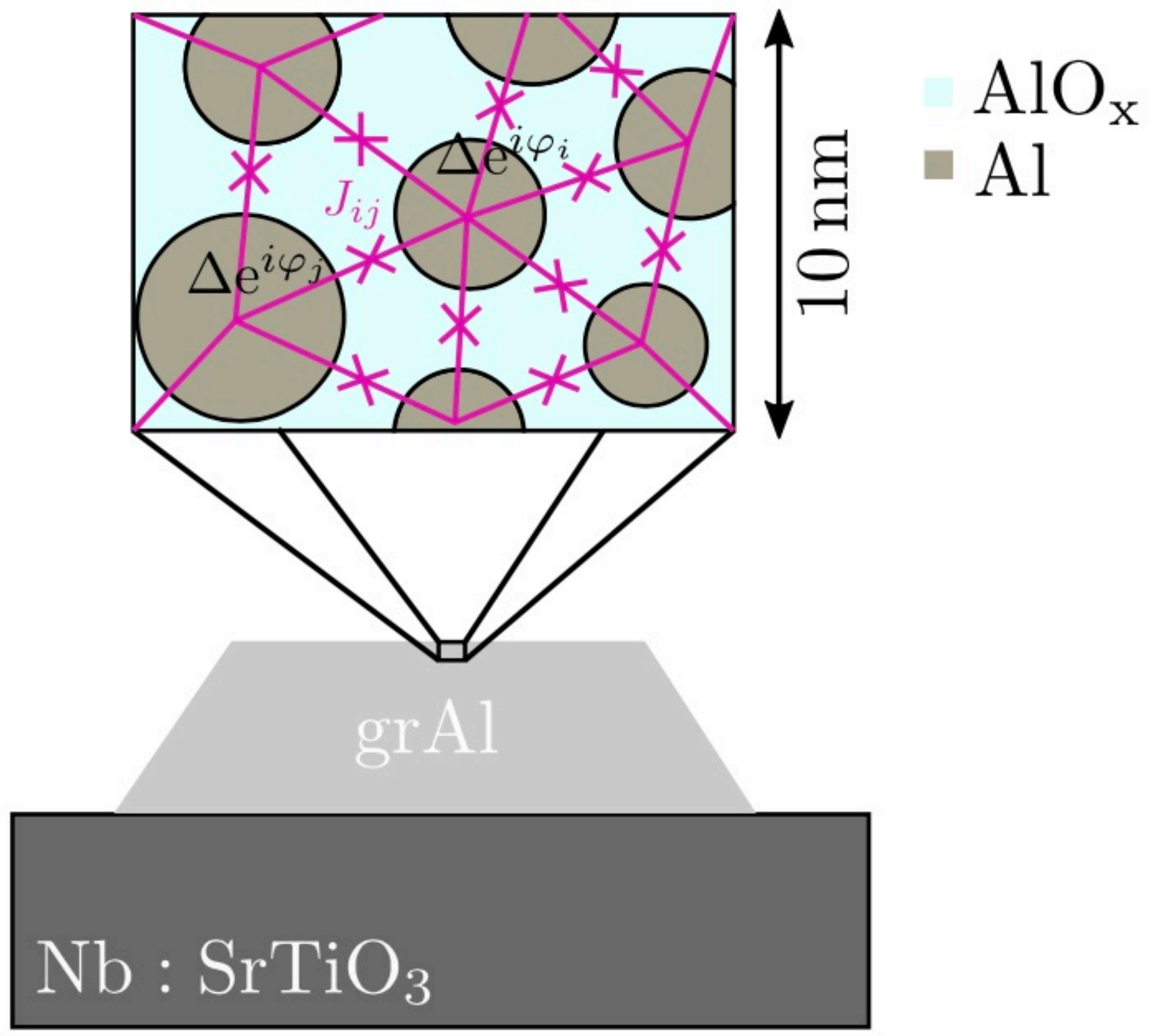
Cooper pair tunneling



- At high currents and with decoupled grains, part of the bias voltage needs to drop between grains
- Combination of Andreev reflection and Josephson tunneling
- Can only be explained, when we take into account the full charge transport

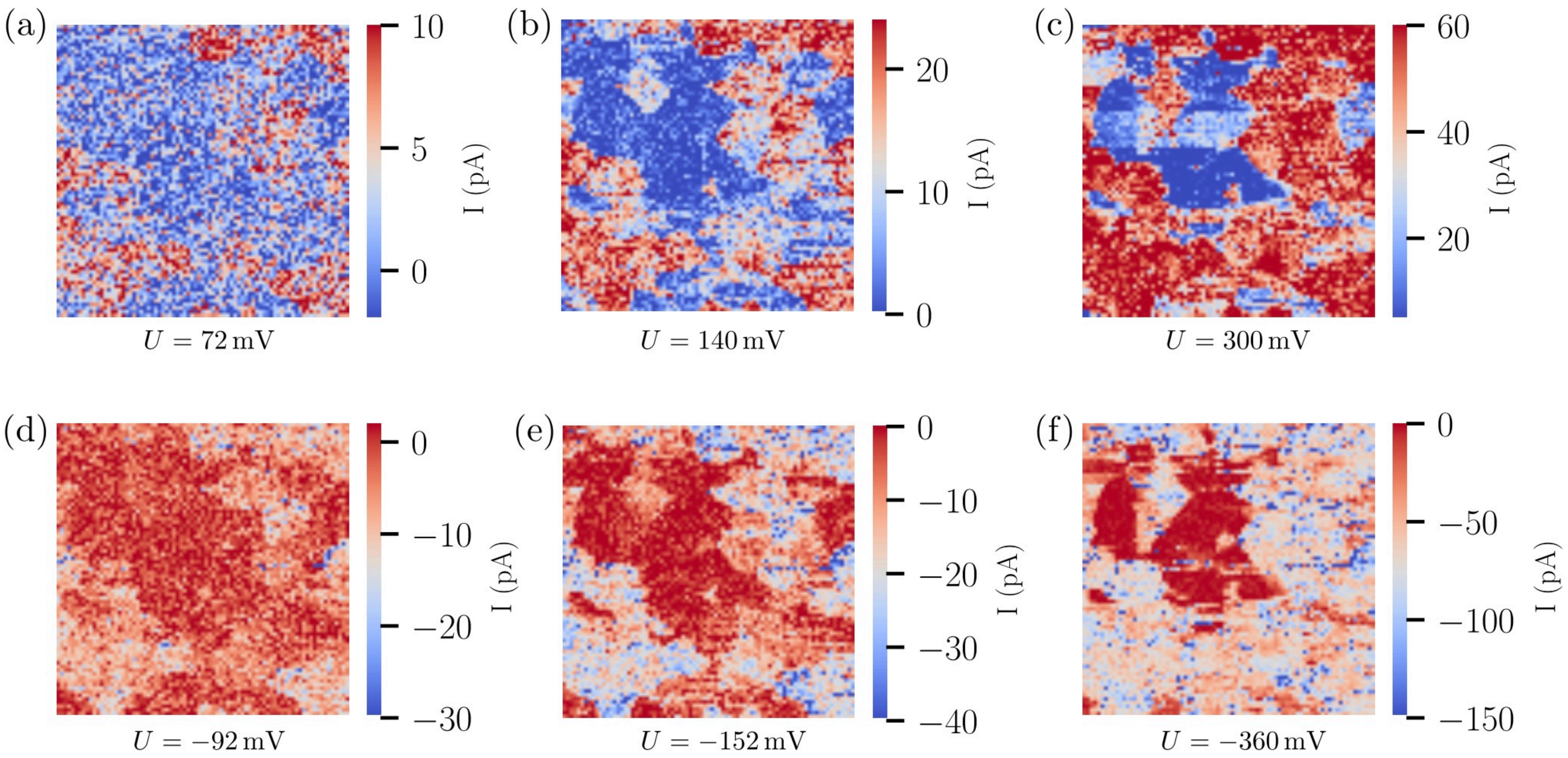
Oxygen rich samples (2000 $\mu\Omega$cm)





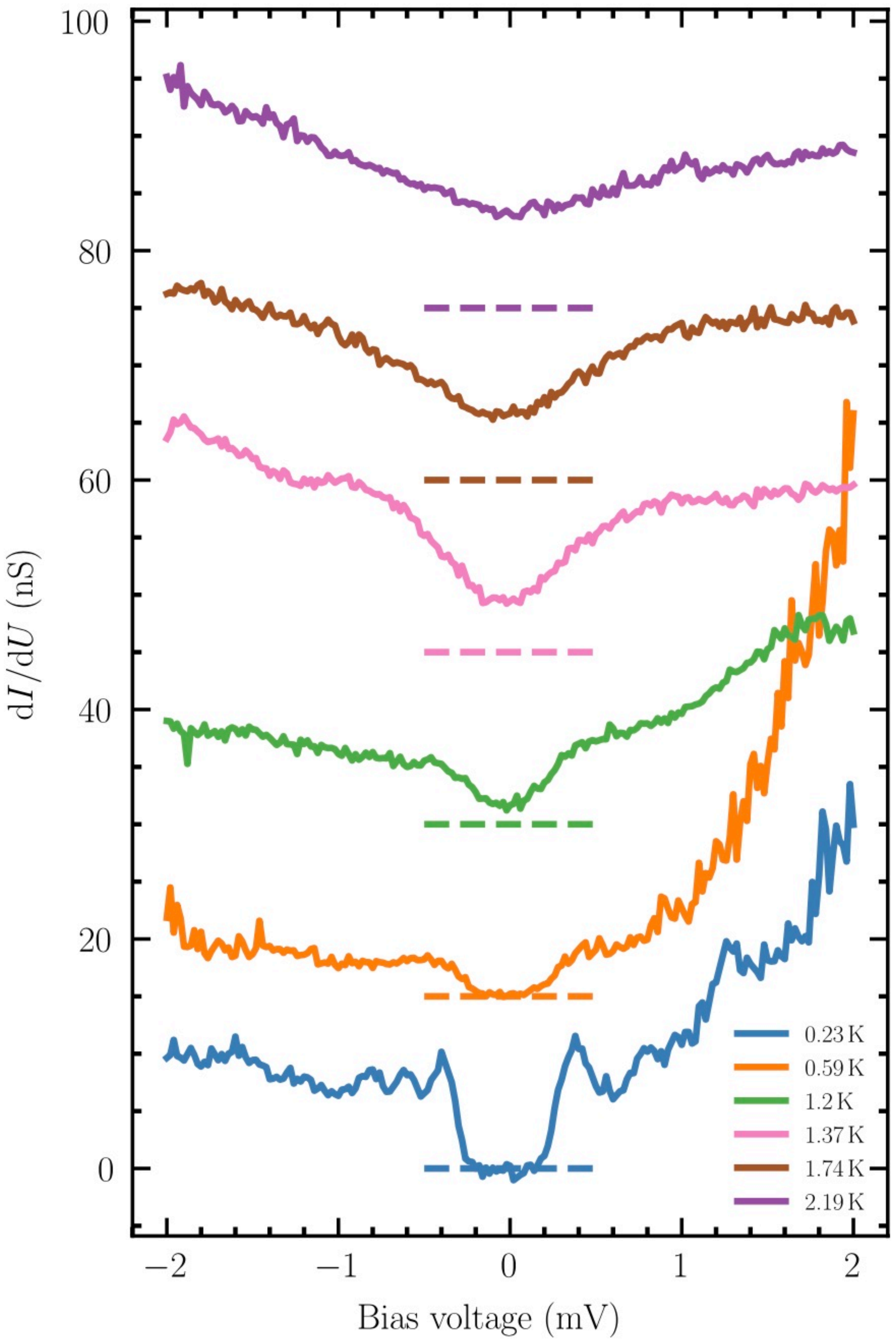
- No relevant change in grain size
- Strong variations from grain to grain in I(V) curve
- Clear charging effects observable
- Grains start to decouple, Coulomb blockade sets in
- Lifetime broadening of charging peaks $\Gamma \gg \Delta$
- Film is not yet insulating

Oxygen rich samples (2000 $\mu\Omega$cm)



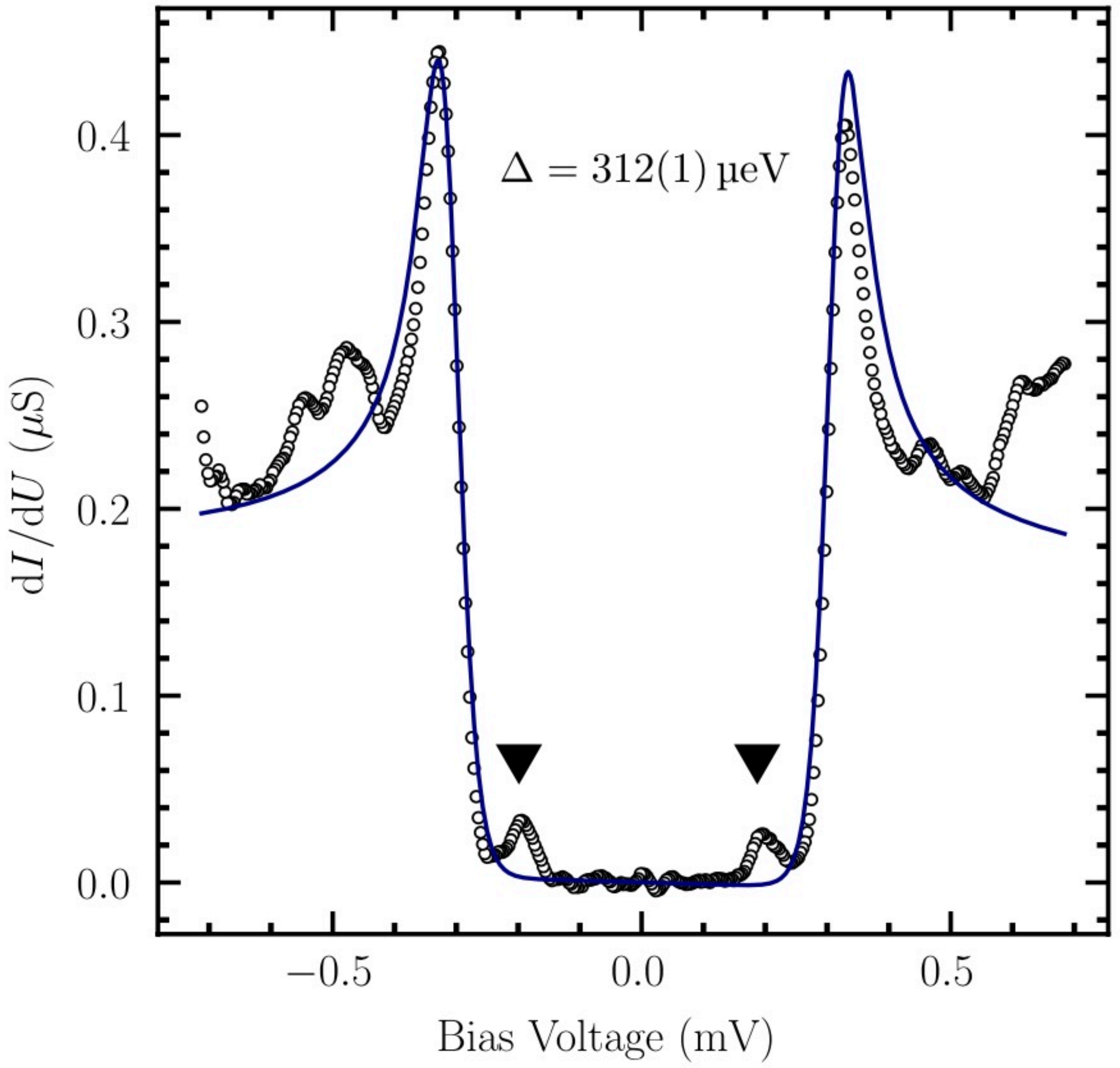
- I(V) maps show charging steps extended over several grains
- With $E_c = \dfrac{e^2}{2C}$ and charging energies between 70 and 300mV, we can estimate capacitances between 0.3-1.1 aF ($\epsilon_r = 9.0$)
- Coulomb clusters of about 10 grains also agrees with recent transmon qubit experiments (Winkel et al. PRX 10,031032)

Oxygen rich samples (2000 $\mu\Omega$cm)

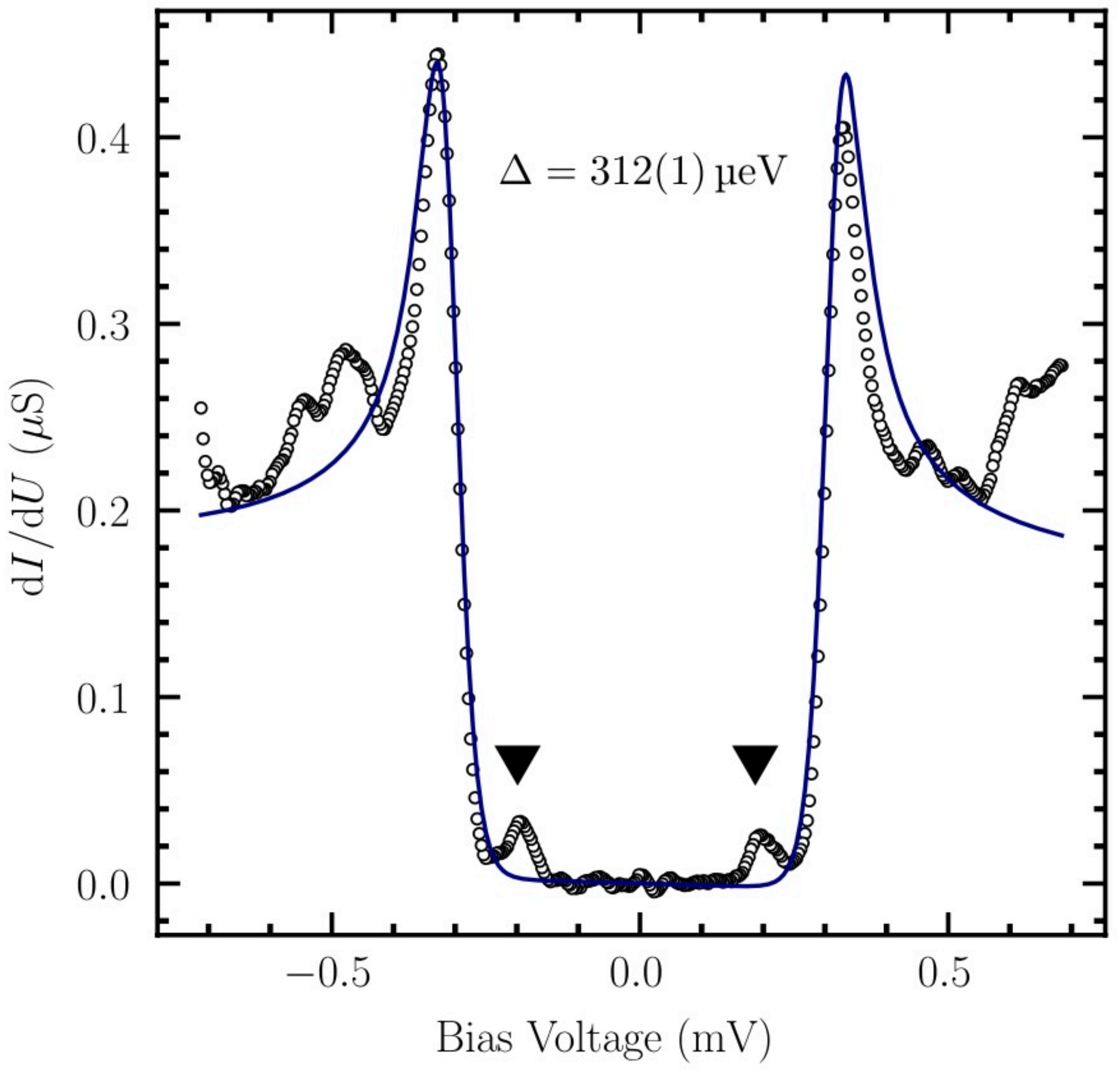


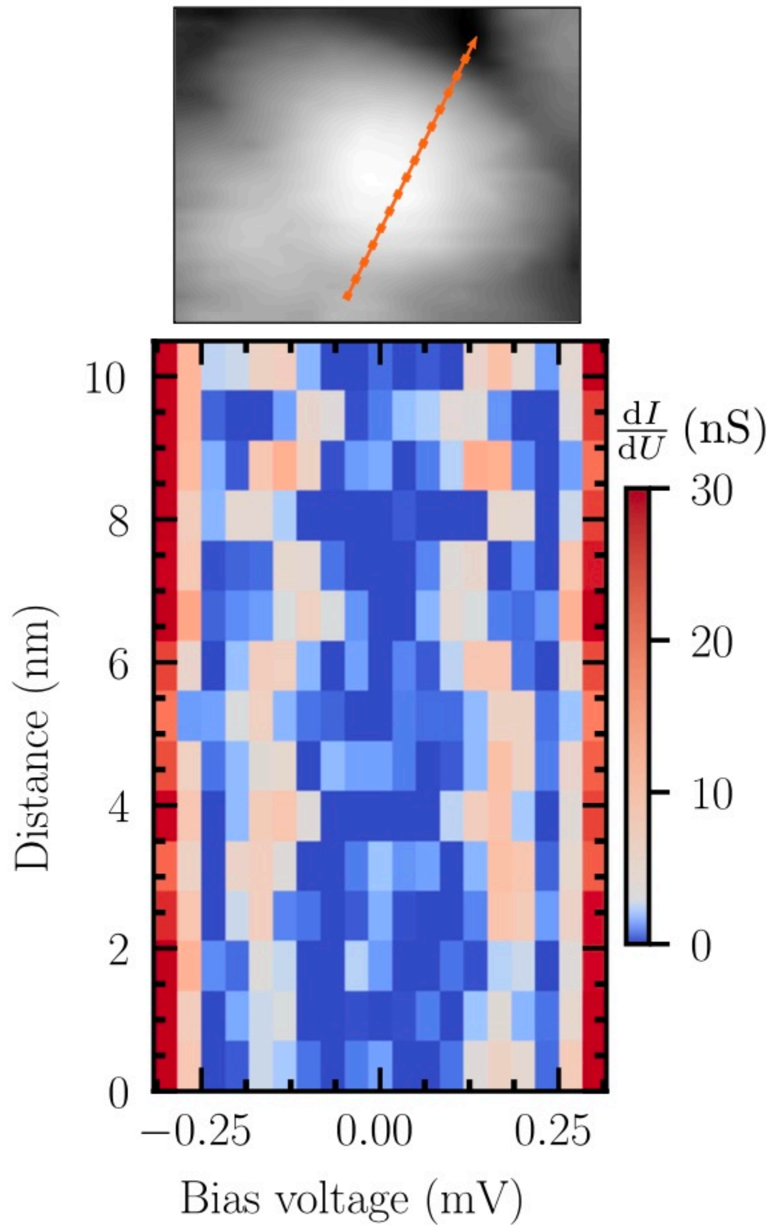
- Sample still shows superconducting gap in low-energy spectrum
- Gaps is even larger than in oxygen poor samples
- Gap in homogeneous
- T$_c$ is much enhanced compared to bulk Al

- In gap states are occasionally found on individual grains
- In gap states can cause decoherence



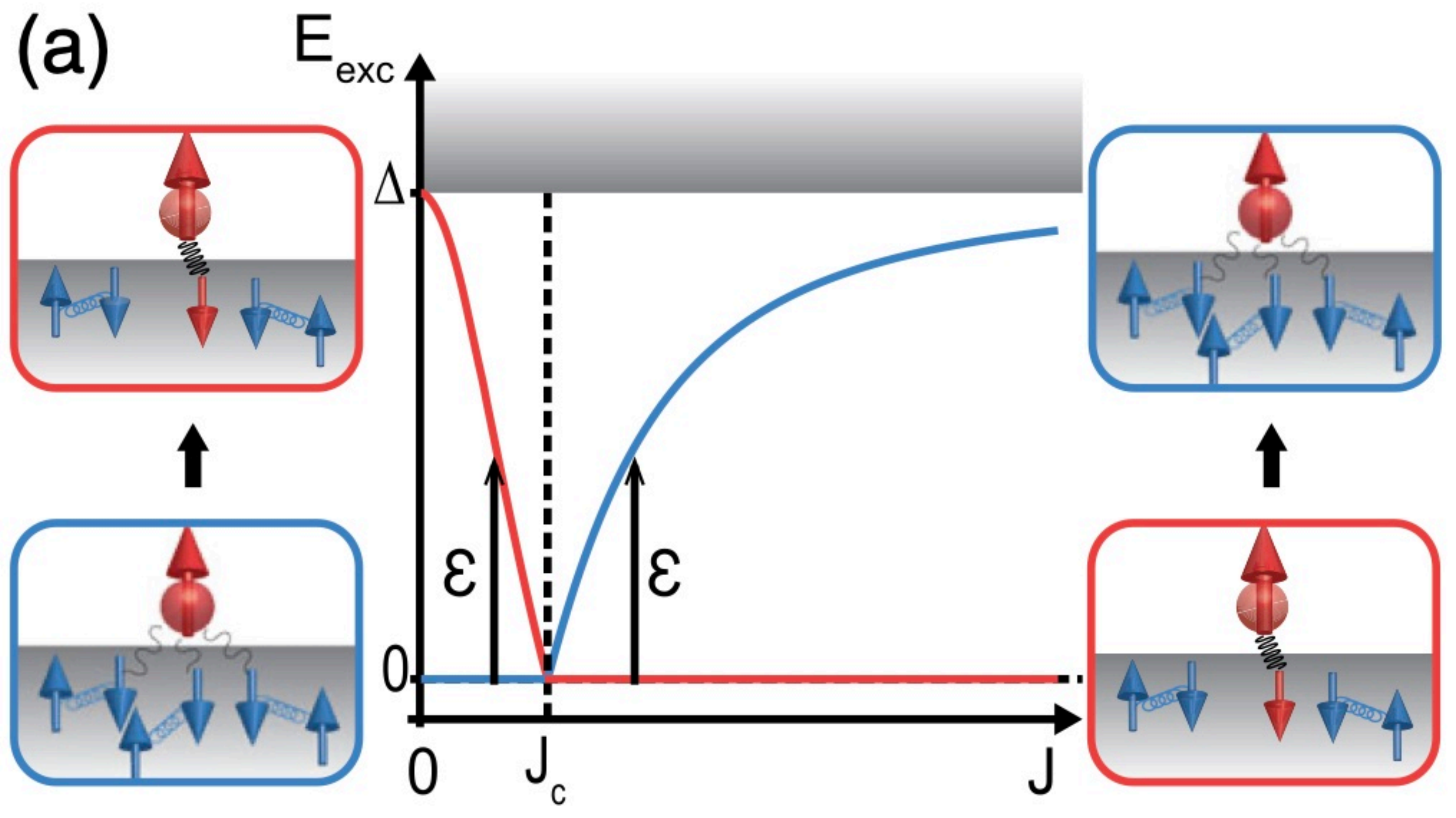
In gap states in oxygen rich samples (2000 $\mu\Omega$cm)





- Sample is (super)conducting, i.e. no carrier localization
- Energy of coherence peak does not change when moving tip → no change of chemical potential of grain due to electric field
- Energy of YSR peak does change
- Imputity spin cannot be located on grain but inside the oxide → unpaired electron in dangling bonds of unordered AlOx

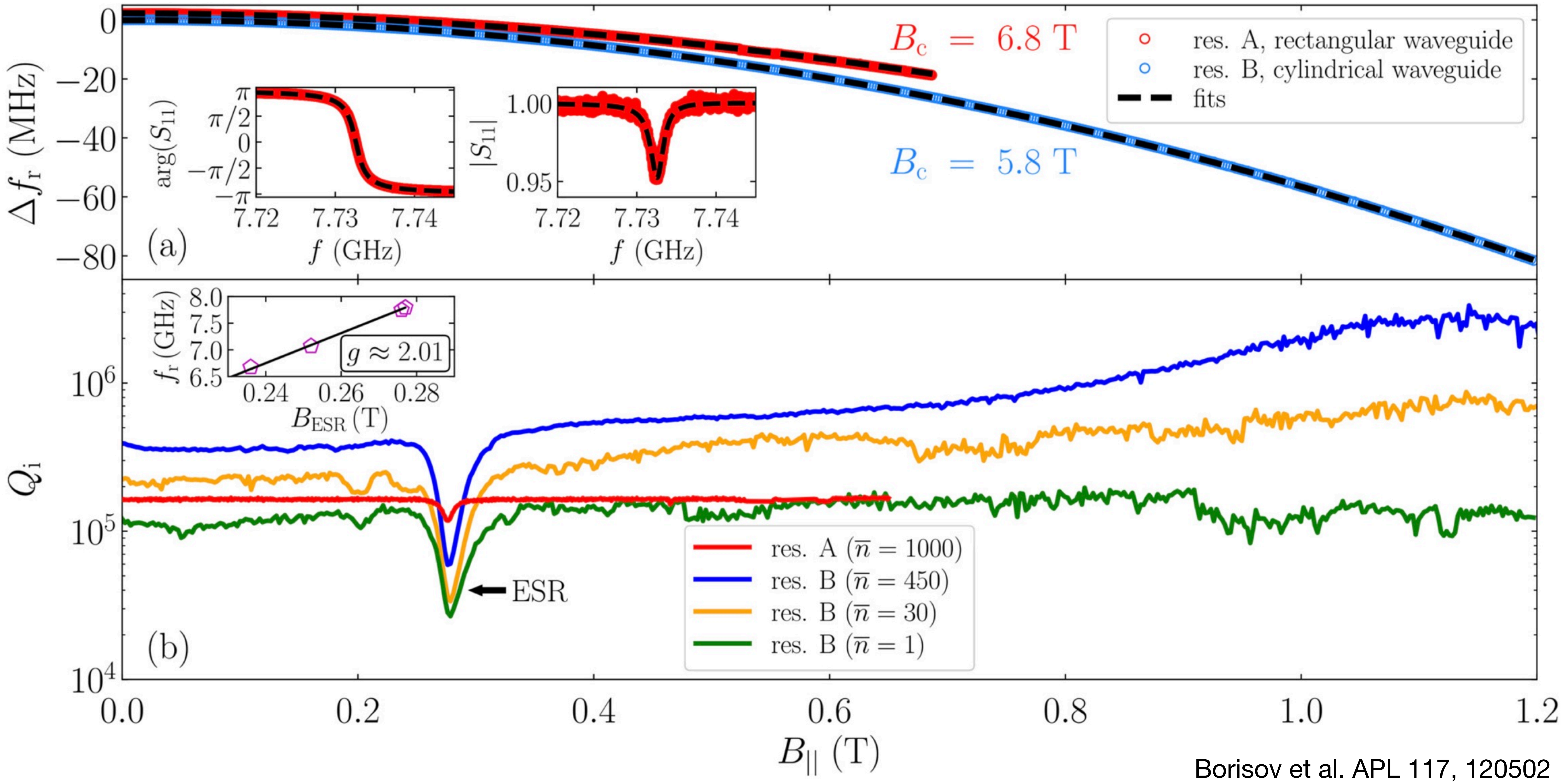
- Speculation of quantized carriers on individual grains, odd number of carriers leads to unpaired electrons
- Unpaired electrons interact with the condensate such that Yu-Shiba-Rusinov states may form
- Competition between pair breaking/screening of the localized spin and paring



Farinacci et al., PRL 121, 196803

- Agrees with Kondo effect near metal-to-insulator transition (see Bachar et al. PRB 87, 214512)

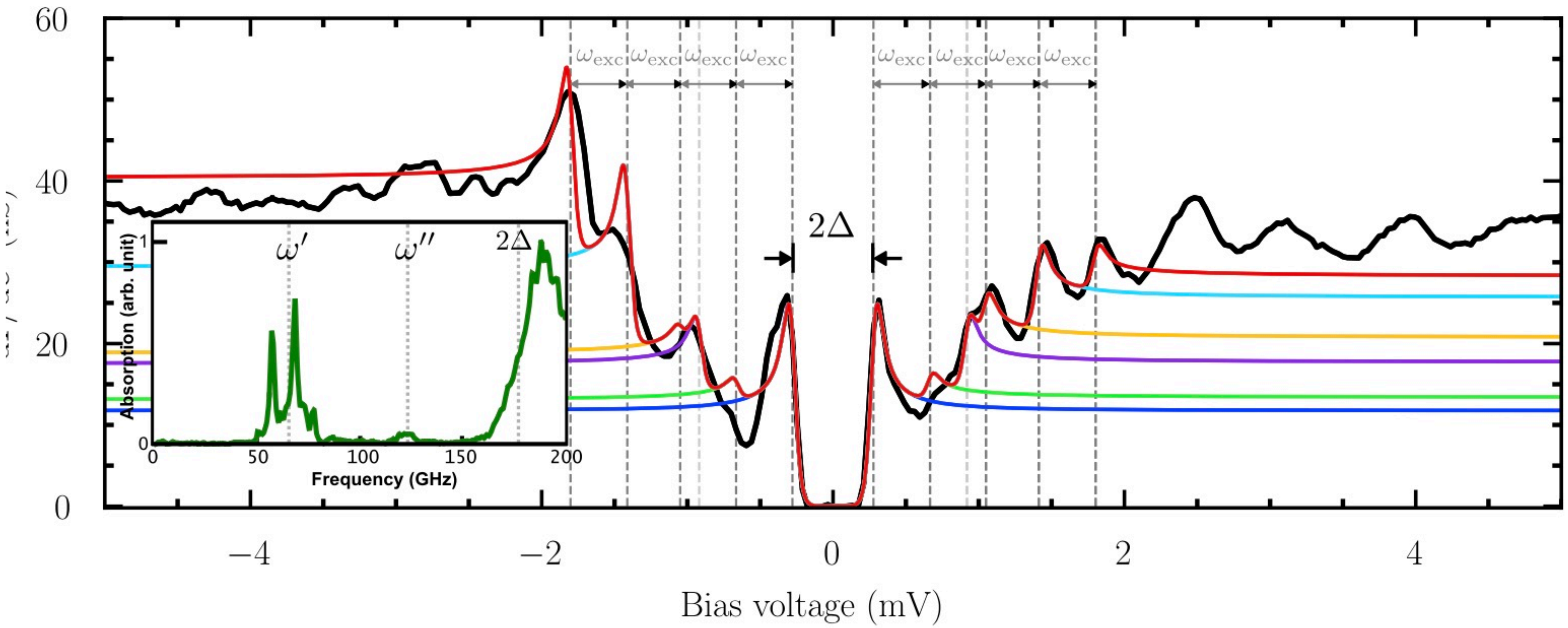
ESR signals in grAl resonators

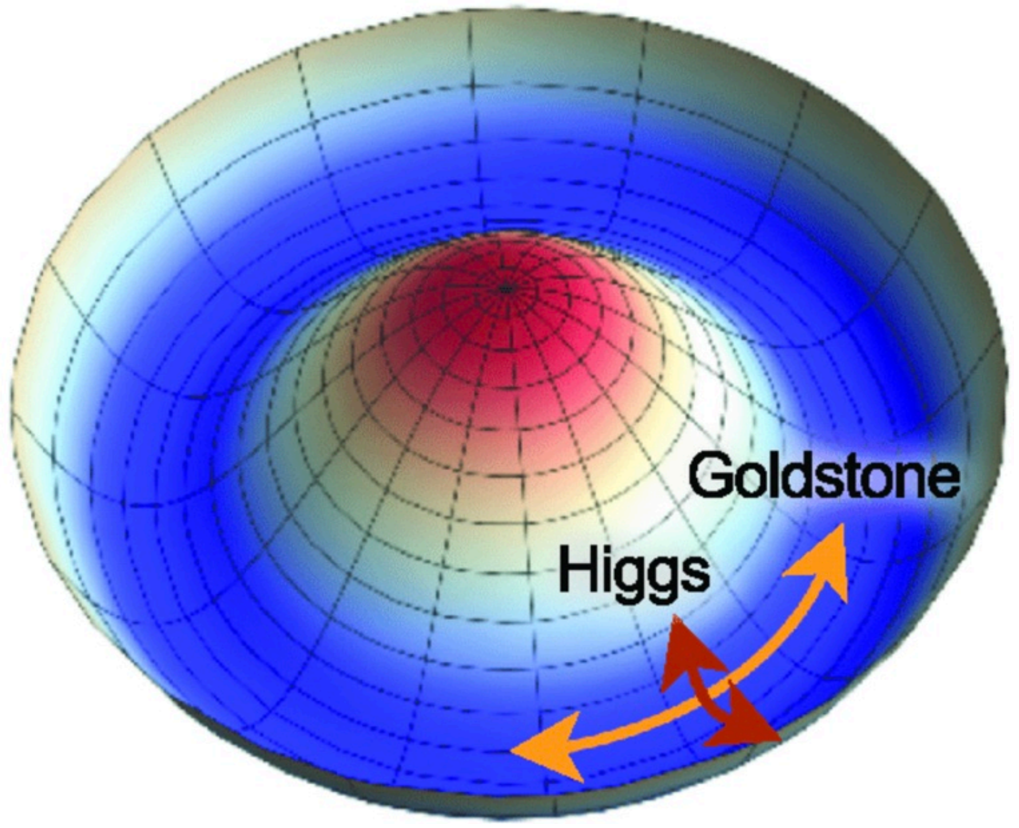


Borisov et al. APL 117, 120502

- Unpaired electrons (s=1/2) with g≈2 were confirmed by ESR signal with field in the plane manifested as drop in quality factor in resonators

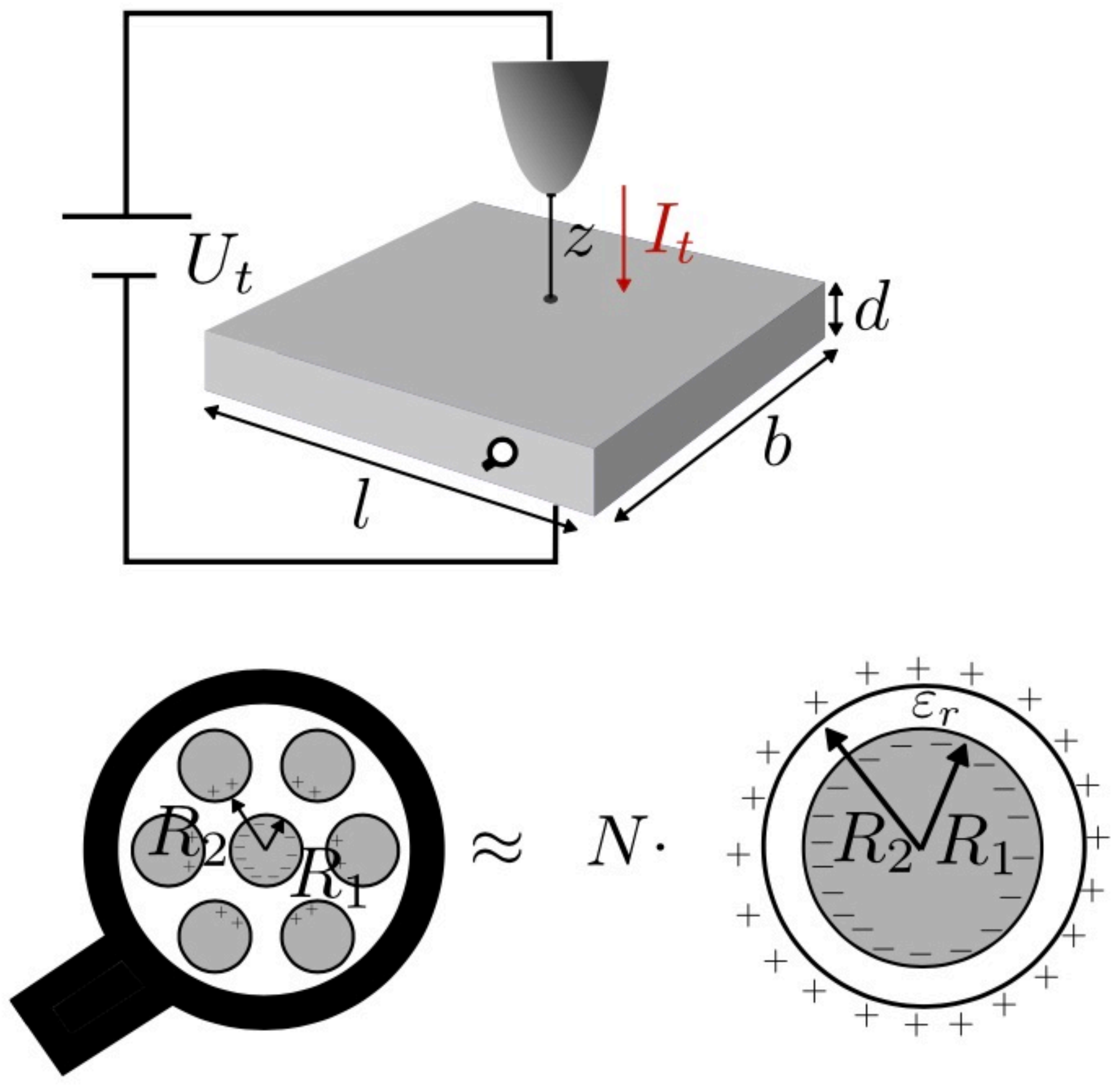
Excitations in oxygen rich samples (2000 $\mu\Omega$cm)

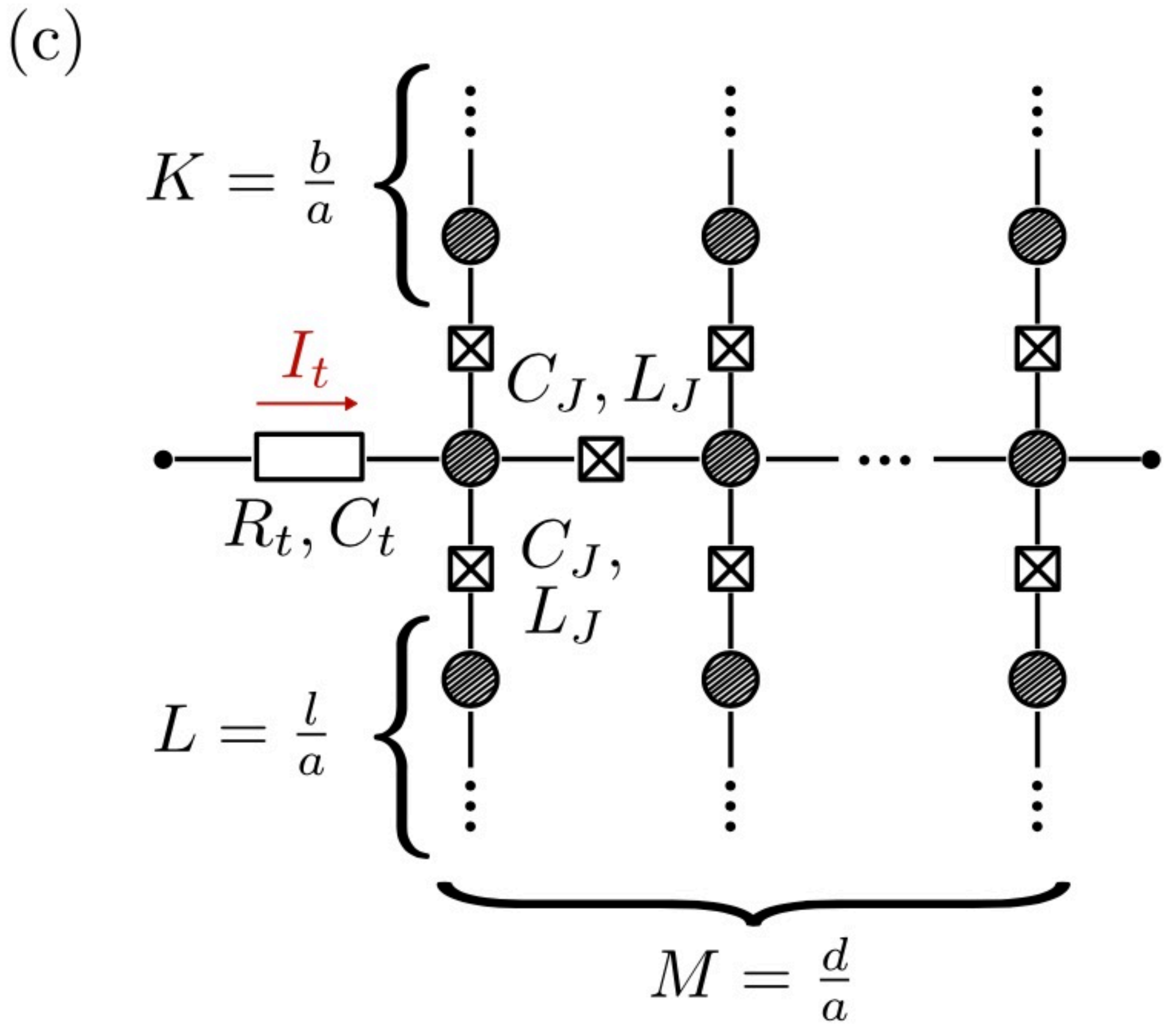




- Nature of the pseudo gap ?
- In oxygen rich samples, clear inelastic excitations exist outside the gap with energies roughly $E = \Delta + n\hbar\omega_{exc}$
- Spectrum can be fitted with up to 5 excitations per tunneling electron
- $\hbar\omega_{exc} \approx 1.4\Delta$ agrees well with THz absorption measurements on grAl samples with same sheet resistance (Levy-Bertrand et al., PRB 99 094506)
- Higgs mode near S-I-transition can be long-lived excitations with $\hbar\omega < 2\Delta$ (Sherman et al. Nature Phys. 11, 118)
- Why only 5 excitations?

Plasmon modes oxygen rich samples (2000 $\mu\Omega$cm)





- LC resonator model

$$C_J = 4\pi\varepsilon_0\varepsilon_r \frac{R_1 R_2}{R_1 + R_2}$$

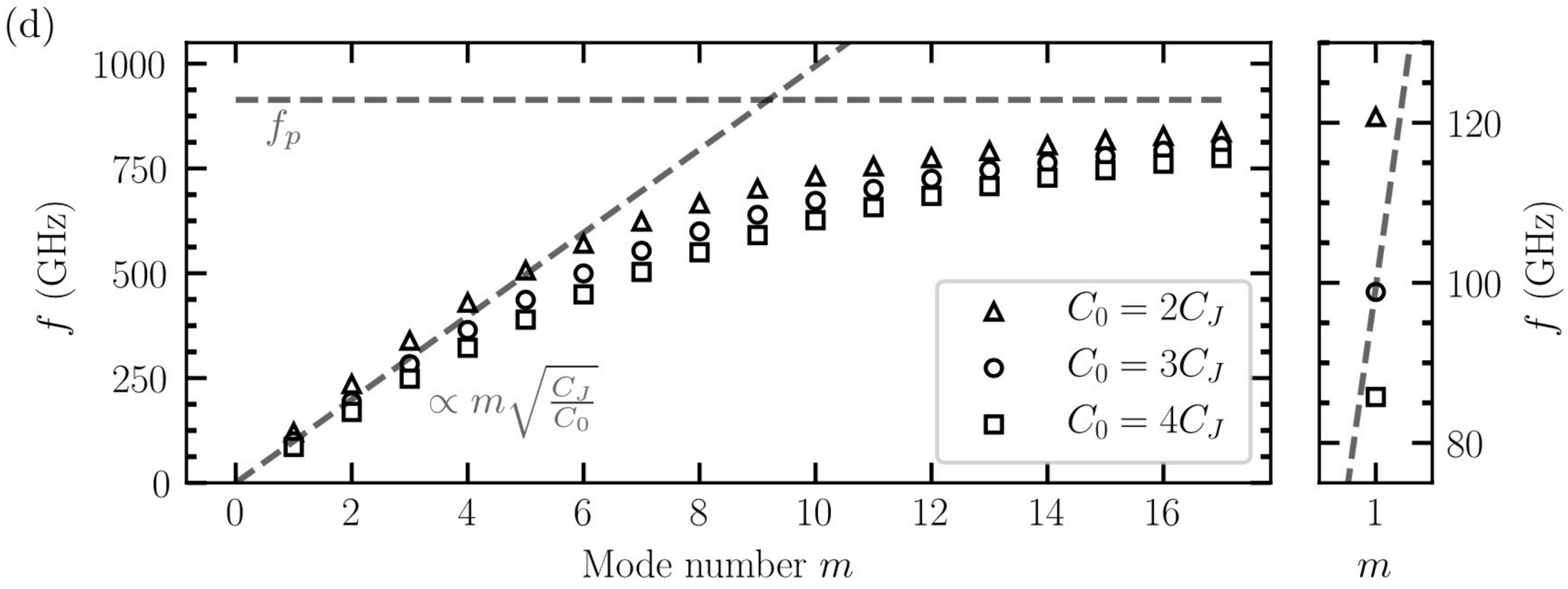
$$L_J = \frac{\Phi_0}{2\pi I_c}$$

R_1 = 1.25 nm R_2 = 1.75 nm C_J = 0.73 aF
j_c = 0.88 mA/$(\mu m)^2$

$$f_p = \frac{\omega_p}{2\pi} = \frac{1}{2\pi}\sqrt{\frac{1}{L_J C_J}}$$

f_c=914 GHz

Plasmon modes oxygen rich samples (2000 $\mu\Omega$cm)



$$\omega_m = \frac{m\pi}{M}\left(L_J\left(C_0 + \frac{m^2\pi^2}{M^2}C_J\right)\right)^{-1/2}$$

- Plasmon modes confined in thin film:

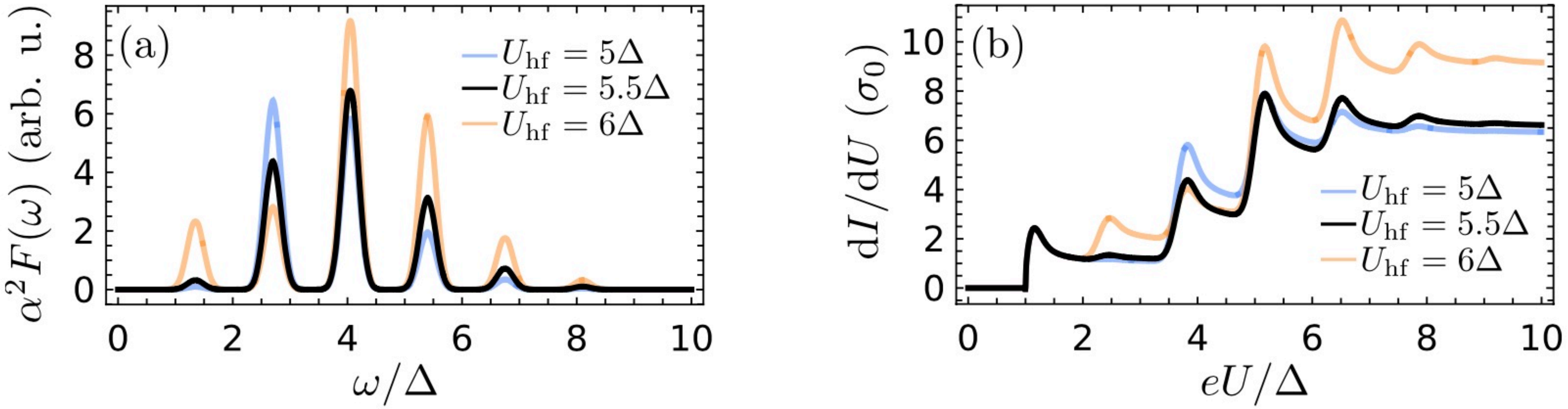
$\rightarrow \omega_1 = 1.28 - 1.8\Delta$

Tien-Gordon Model for spontaneous plasmon emission

$$
\begin{aligned}
\sigma_{\mathrm{s}}^{\mathrm{tot}}(eU) &= \int\limits_0^{eU} \mathrm{d}\omega\, \sigma_{\mathrm{s}}^{\mathrm{el}}(eU-\omega) \sum_n J_n^2\left(\frac{eU_{\mathrm{hf}}}{\omega_{\mathrm{exc}}}\right) \delta_a(\omega - n\omega_{\mathrm{exc}}) \\
&= \sigma_{\mathrm{s}}^{\mathrm{el}}(eU) + \int\limits_0^{eU} \mathrm{d}\omega\, \sigma_{\mathrm{s}}^{\mathrm{el}}(eU-\omega) \underbrace{\sum_{n=1} J_n^2\left(\frac{eU_{\mathrm{hf}}}{\omega_{\mathrm{exc}}}\right) \delta_a(\omega - n\omega_{\mathrm{exc}})}_{\propto \alpha^2 F(\omega)}
\end{aligned}
$$

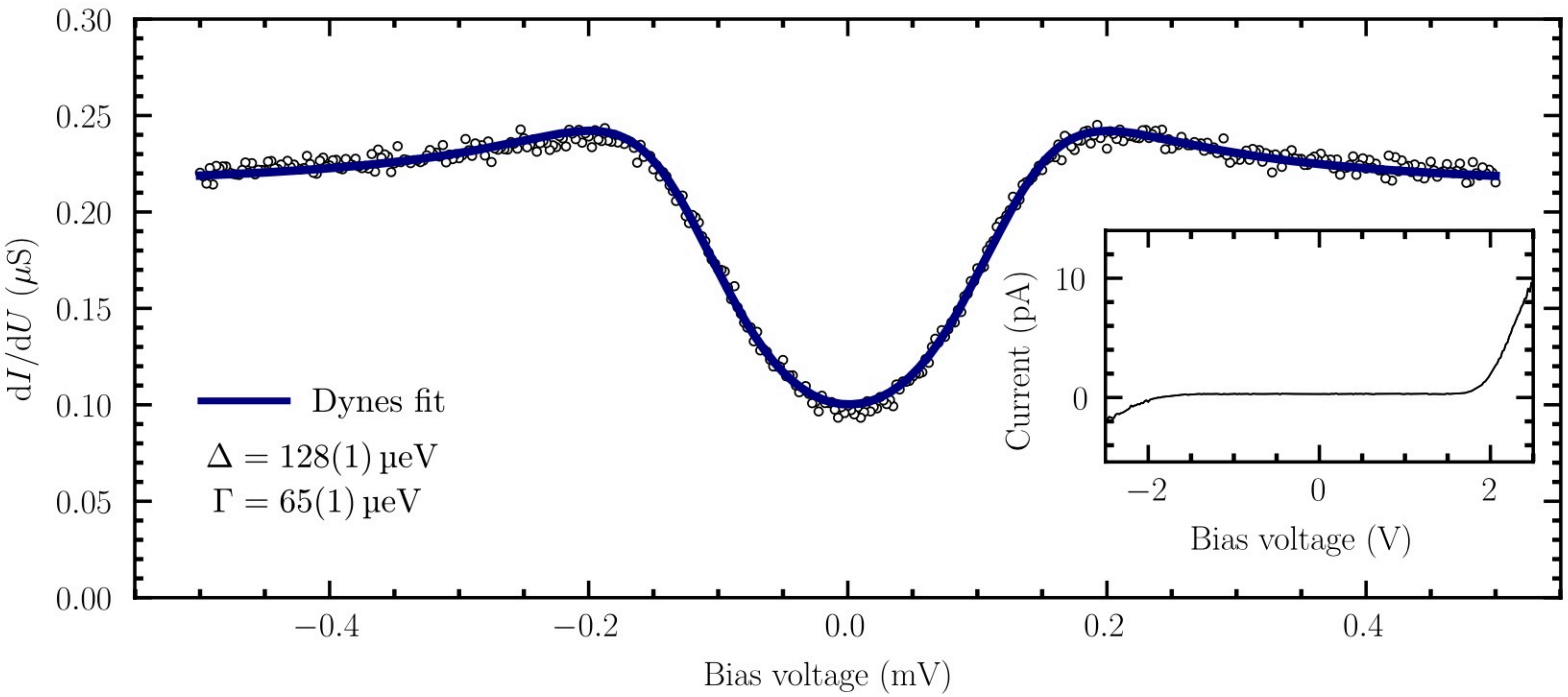
with

$$\delta_a = \frac{1}{|a|\sqrt{\pi}} \mathrm{e}^{-(x/a)^2}.$$



- Tien-Gordon model naturally explains intensities of excitations

AlOx on single crystal Al(111)



- Preliminary results!

- Oxydation in UHV with 2.7 x 10^4 L, corresponds to 2 ML or 0.8 nm

- Clearly, the insulating oxide is detected as an insulating barrier in I(V) curves
- Incomplete oxidation, many dangling bonds
- Local DOS very smeared, reduced gap (128 versus 166 μeV) and short life time (Dynes fit)
- Oxide barrier measured locally is far from ideal insulator

Thank you for your attention!

Questions ?