Absence of zero-energy surface bound states in Cu<sub>x</sub>Bi<sub>2</sub>Se<sub>3</sub> via a study of Andreev reflection spectroscopy

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### Main Motivation:

# Search of a realization of Topological Superconductor

### <u>Topological Superconductor (TSC) :</u>

Superconducting state with a full bulk gap protected by particle-hole symmetry, but with stable gapless (Majorana) surface bound states. (candidates: <sup>3</sup>He-B phase; interface topological insulator/s-wave SC; *p*-wave SC)

## analog to:

# <u>Topological Insulator (TI)</u>:

Insulating state with a bulk gap protected by time-reversal symmetry, but with gapless surface states with an odd number of Dirac points. (example:  $Bi_2Se_3$ )

-[2010]: superconductivity below  $T_c = 3.8$  K was observed in an electrondoped topological insulator  $Cu_x Bi_2 Se_3$  [Y. S. Hor et al., Phys. Rev. Lett. **104**, 057001]

-[2010]: Theory Proposed  $Cu_xBi_2Se_3$  as candidate of TSC using a phenomenological model, predicting the existence of an odd-parity full SC gap with TRS and a resultant zero-energy surface Andreev bound states (Majorana fermions) [L. A. Fu, and E. Berg, Phys. Rev. Lett. **105**, 097001 (2010)]

-[2011]: This appears to be confirmed by the successful detection of a zero-bias-conductance-peak (ZBCP), as evidence for the existence of zero-energy Majorana fermions, by means of a "soft" point-contact technique. [S. Sasaki, M. Kriener, K. Segawa, K. Yada, Y. Tanaka, M. Sato, and Y. Ando, Phys. Rev. Lett. **107**, 217001 (2011)]



In view of the significance of the observation and the complexity involved in point-contact devices, further **stringent** experimental test on the existence of zero-energy Majorana fermions is needed. A newly developed nanoscale Andreev reflection (AR) spectroscopy method employed to elucidate the superconducting gap structures of  $Cu_xBi_2Se_3$  as a function of temperature and magnetic field.

The results show that the ZBCP can be tuned in or out from  $Cu_xBi_2Se_3$  samples depending on the normal metal/superconductor (N-S) barrier strength *Z*.

"While the appearance of ZBCP may be traced to different origins, its **absence under finite barrier strength entails the absence of zero-energy Majorana fermions**"

(the ZBCP should be persistently present in all N-S junctions with different barrier strength Z ranging from the transparent limit to the tunneling limit).

"The present observations thus call for a reexamination of the nature of the superconducting state in Cu<sub>x</sub>Bi<sub>2</sub>Se<sub>3</sub>".

Single crystals of  $Cu_xBi_2Se_3$ , with x=0.15, up to cm size with *c*-axis preferred. Measured an onset bulk  $T_c$ =3.4 K and a superconducting volume fraction up to =20 % at 2 K (close to results previously reported).

Newly developed technique to construct nanoscale N-S devices and perform Andreev reflection spectroscopy:

- The bulk sample of Cu<sub>x</sub>Bi<sub>2</sub>Se<sub>3</sub> is mechanically cleaved into micro-scale crystals;
- Immediately transferred into a vacuum chamber;
- A sharp probe tip attached on a micromanipulator is used to place a piece of microcrystal on top of multiple parallel metal electrodes spaced ≈500 nm apart (as designed via electron-beam lithography)



• To obtain AR spectra for a target N-S junction, a special circuit is used, where a small AC current superimposed to a DC bias current is applied between the I<sub>+</sub> and I<sub>-</sub> terminals, while both the DC and the AC voltages across the N-S junction are measured between the V<sub>+</sub> and V<sub>-</sub> terminals.



The four central electrodes (labeled as 2-5) are designed to be 1  $\mu$ m wide while the two external electrodes (labeled as 1 and 6) are 4  $\mu$ m wide.

The flexibility in selecting the configuration for the measurement terminals enables one to study various N-S junctions between different metal electrodes and the same superconductor crystal with **different** Z **parameters** in a single device.



• Three different N-S junctions;

• T = 240 mK;

- *dI/dV* dip at zero bias;
- Two shoulders at  $V \approx \pm 0.4 \text{ mV}$ ;
- Typical for N-S interface with finite barrier strength *Z* according to the BTK theory;
- Solid lines represents a fitting of the experimental *dI/dV* data (normalized to the normal state data) by the generalized BTK theory, including broadening;
- Considering the coexistence of SC and non-SC phases in bulk  $Cu_xBi_2Se_3$ , one can express the total normalized conductance as:  $\sigma = w\sigma_s + (1 - w)$
- From the fitting one gets the gap energy:  $\Delta = 0.35 \pm 0.04 \text{ meV};$   $\Gamma = 0.13 - 0.27 \text{ meV}$

<u>Note 1</u>: The three junctions have different values of w (from 13% to 80%), different values of Z, but they all notable yield the same value for the gap  $\Delta$ .

<u>Note 2</u>: These three "spectroscopic" junctions have rather large normal state resistance  $R_N \gg 100 \Omega$ , whereas for junctions with small  $R_N$  ( $\leq 10 \Omega$ ) the behavior is different:

• *dI/dV* zero-bias peak;

- followed by a dip at the peak edge;
- typical for nearly transparent N-S interface with weak barrier strength;



Large- $R_N$  junctions ( $\gg 100 \Omega$ )

No ZBCP Lateral Peaks (shoulders)

Good indication of the gap  $\Delta$ 

Low- $R_N$  junctions ( $\leq 10 \Omega$ )

ZBCP Lateral Dips No information on the gap  $\Delta$ 

Observations consistent with the commonly adopted empirical formula for determining ballistic transport regime (  $a \ll \ell$  ):

 $R_{\rm N} \gg 4\rho/3\pi\ell$ 

(with  $\rho$  bulk material resistivity and  $\ell$  mean free path in the junction).

Meaning:  $R_{\rm N} = (4\rho\ell)/(3\pi a^2)$ 

the point contact size has to be less than the mean free path to ensure ballistic transport across the N-S interface, which is sufficient for providing correct spectroscopic, energy-resolved information of the superconducting gap.

In this system ( $\rho \approx 140 \ \mu\Omega cm$ ,  $\ell \approx 45 \ nm$ ) the rule gives a critical  $R_N \approx 13 \ \Omega$ .

compatible with phenomenology

The zero bias peak for the transparent junctions can be explained as the Andreev reflection plateau within the bulk superconducting gap, with a cut-off of plateau width corresponding to reaching the critical current at relatively low bias V <  $\Delta$ /e because of the low junction resistance.

The width of the ZBP does not reflect the gap energy correctly, because the normal state resistance  $R_N$  is less than the threshold  $R_N$  value for ballisticity ( $\approx 13 \Omega$ ), and thus these junctions are scattering electrons: the estimated point contact size is larger than the electron mean free path.

(Threshold case of  $R_N \approx 13 \Omega$  has a ZBP width compatible with the energy gap.) Additional check: same low-resistance junctions measured again after 6 months, exhibit larger  $R_N \approx 100 \Omega$  and compatible extracted gap values.

### Magnetic field evolution:

# Large-R<sub>N</sub> junctions ( $\gg 100 \Omega$ )

- The double peaks move to lower bias and decrease in amplitude;
- Double-peak feature disappears completely at  $B \approx 5$  T, close to the nominal upper  $B_{c2}$ ;
- Double dips at high bias move towards the center in a much faster pace;

# Low- $R_N$ junctions ( $\leq 10 \Omega$ )



- The zero-bias conductance peak is already greatly suppressed at B = 3 T;
- The zero-bias conductance peak disappears at B = 5 T;
- *dI/dV* oscillations supra-gap but before the reach of the normal state;

### Comparison with previous experiment:



- The ZBCP is on top of a strong nonsaturating V-shape *dI/dV* background;
- Observed ZBCP disappears completely in a magnetic field  $\approx 0.45$  T (an order of magnitude smaller than  $B_{c2}$ );



- Reproduced similar behaviors by placing the SC microcrystals onto the electrodes while still electrically disconnected;
- Electrical connection established via annealing (short voltage pulse);
- Left: similar features, ZBP destroyed at B ≃0.4 T, indicating a thick degraded SC surface layer dominating the N-S transport
- Right: pure strong V-shape spectrum, no hint for AR signature, can be explained by a thick non-superconducting surface layer.

### Cross check:

For two additional junctions in the same device, which were electrically connected initially without the need of voltage annealing, the AR spectra are dominated by a pronounced ZBCP but with much smaller V-shape background (likely due to much less contribution from a non-SC surface layer), and the ZBCP is suppressed at higher B field  $\approx 2$  T (indicating better-quality SC layers at the junctions).





### Conclusion:

"Therefore, our control experiments suggest that a ZBCP disappearing at a magnetic field much lower than the bulk critical field and showing a strong non-saturating V-shape background reflects the existence of a **degraded** superconducting surface layer in **non-ideal** N-S junctions, and thus **cannot be used as the signature for zero-energy Majorana fermions**".