Production of nonlocal quartets in a Josephson bijunction and nonlocal transport beyond the crossed Andreev reflection current

> Régis Mélin Institut NÉEL Grenoble France



Organization of the Talk

- 0. Introduction on crossed Andreev reflection (1S, 2N, 2 voltages, 0 phase)
- 1. Nonlocal quartets in metallic systems (3S, 1 voltage, 1 phase)
 - 1.a. Phenomenology, microscopic process, π -junction
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SN Junction: Andreev Reflection (1/3)



SN Junction: Andreev Reflection (2/3)



SN Junction: Andreev Reflection (3/3)



SN Junction: Nonlocal Andreev Reflection (1/3)



Nonlocal Andreev Reflection (2/3)



Nonlocal Andreev Reflection \equiv Cooper Pair Splitting (3/3)



Three-terminal set-up required in experiments First theoretical contributions: Byers-Flatté, Martin, Anatram-Datta, Deutscher-Feinberg, Falci-Hekking, Choi-Bruder-Loss, Mélin



Structure: Superconductor + ferromagnetic electrodes



Voltage on one contact as a function of current through the other contact: nonlocal resistance



High transparency \rightarrow large signal



Nonlocal resistance depends on relative spin orientation \rightarrow Magneto-resistive effects

Beckmann, Weber, von Löhneysen, PRL '04



FIG. 2 (color online). Nonlocal resistance *R* as a function of the temperature *T* at zero external magnetic field for two contacts of sample *T2* at a distance d = 310 nm. The solid and dashed lines correspond to parallel and antiparallel alignment of the injector and detector magnetizations, respectively. Inset: nonlocal resistance *R* at *T* = 1.8 K as a function of a magnetic field *B* applied parallel to the iron wires, for two contacts of sample S5 at a distance d = 210 nm. The resistance jumps correspond to magnetization reversals of the ferromagnetic contacts. The arrows indicate the four different magnetization states.

197003-2



FIG. 4 (color online). Difference $\Delta R_{\rm NS}$ between parallel and antiparallel alignment vs distance *d* between the contacts, for two samples in both the normal (open symbols) and superconducting (closed symbols) state. The solid and dashed lines are fits to (1) and (5), respectively, as described in the text.

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Perturbation Theory in the Number of Nonlocal Green's Functions R. Mélin and D. Feinberg, PRB '04

Small parameter of perturbation theory $\sim \exp(-2R/\xi)$



FIG. 2. Variation of the normalized linear crossed conductances $\lambda G_{\rho}(P)$ (solid line) and $\lambda G_{d\rho}(P)$ (dashed line) as a function of the spin polarization *P*, for τ =0.25 (a), τ =0.5 (b), τ = =0.75 (c), τ =2 (d). Note the different scaling factors on the conductance axis, and τ =1.

First evidence for negative nonlocal conductance in a highly transparent NSN structure (P = 0)

Régis Mélin, Institut NEEL, Grenoble

Production of nonlocal quartets in a Josephson bijunction

Quasiclassics: General Solution in Ballistic Limit M.S. Kalenkov and A.D. Zaikin, PRB '07



Two Recent Equivalent Experiments with Quantum Dots (1/2)



Carbon nanotube as an electron beam splitter

Two Recent Equivalent Experiments with Quantum Dots (2/2)



Quantum wire as an electron beam splitter

Two Recent Equivalent Experiments with Quantum Dots (2/2)



Experimental signatures for separated electron pairs $|1\uparrow;2\downarrow\rangle$

Two Recent Equivalent Experiments with Quantum Dots (2/2)



These experiments do not probe directly entanglement, for instance $(|1\uparrow; 2\downarrow\rangle - |1\downarrow; 2\uparrow\rangle)/\sqrt{2}$

Spectroscopy (theory) Byers and Flatté, PRL '95



Metallic contact + STM tip

Spectroscopy (theory) Byers and Flatté, PRL '95



Propagation of quasi-particles in the directions where $\Delta_{\mathbf{k}}$ has nodes

Spectroscopy (theory) Byers and Flatté, PRL '95



Probe of anisotropy of order parameter (High-Tc superconductors and correlated materials)

Summary

- Magneto-resistive effects
- Correlated pair of electrons
- Spectroscopy

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What Theory Brings Into These Problems

- Suggestion of experiments
- Semi-quantitative simulation of set-ups
- Discriminate the features of pair splitting from other effects

Measurement of Current Fluctuations B. Kaviraj, O. Coupiac, H. Courtois, F. Lefloch, PRL 2011



SQUID-based amplifiers

Recursive Green's functions

Scattering calculations

$S_{a,b} > 0$ at high transparency not due to Cooper pair splitting

CEA-Grenoble (Lefloch et al.) NEEL-Grenoble (Mélin et al. '08, '10, '13)

Karlsruhe (Golubev and Zaikin '10)

Current Noise $S_{a,a}$ and Current Noise Cross-Correlations $S_{a,b}$

$$S_{a,a}(t') = \langle \delta \hat{l}_a(t+t') \delta \hat{l}_a(t) \rangle$$

$$S_{a,b}(t') = \langle \delta \hat{l}_a(t+t') \delta \hat{l}_b(t) \rangle$$

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Production of Nonlocal Quartets, $R < \xi$ A. Freyn, B. Douçot, D. Feinberg, R. Mélin, PRL 2011, NÉEL / LPTHE



Intuitively

2 pairs in coherence volume within time interval \hbar/Δ \rightarrow production of a correlated pair of pair between S_a and S_b

Production of Nonlocal Quartets, $R < \xi$ A. Freyn, B. Douçot, D. Feinberg, R. Mélin, PRL 2011, NÉEL / LPTHE



ac Josephson current of pairs from S_a to S and from S to S_b

Production of Nonlocal Quartets, $R < \xi$ A. Freyn, B. Douçot, D. Feinberg, R. Mélin, PRL 2011, NÉEL / LPTHE



ac Josephson current of pairs from S_a to S and from S to S_b

But possibility of a dc Josephson current of quartets
from S to S_a and S_b
if
$$V_a = -V_b$$
 and $V_S = 0$ because $\Delta E = 2e(V_a + V_b - 2V_S) \equiv 0$

Production of Nonlocal Quartets, $R < \xi$ A. Freyn, B. Douçot, D. Feinberg, R. Mélin, PRL 2011, NÉEL / LPTHE



Adiabatic model

$$\phi_{a}(t) + \phi_{b}(t) - 2\phi_{S} = \left[2e(V_{a} + V_{b} - 2V_{S})/\hbar\right]t + \phi_{a} + \phi_{b} - 2\phi_{S}$$

$$I_{quartet}(t) = I_{c}\sin[\phi_{a}(t) + \phi_{b}(t) - 2\phi_{S}]$$

- AC Josephson effect of quartets in general
- DC Josephson effect of quartets if $V_a = -V_b$ and $V_S = 0$










Nonlocal Quartets, DC if $V_a = -V_b$, $V_S = 0$ Quartet Resonance in DC Current for $V_a = -V_b$ A. Freyn, B. Douçot, D. Feinberg, R. Mélin, PRL 2011, NÉEL / LPTHE









Split pair

$$\frac{1}{\sqrt{2}} \left(c^+_{a,\uparrow} c^+_{b,\downarrow} - c^+_{a,\downarrow} c^+_{b,\uparrow} \right)$$
Unstable

$$\frac{1}{2}\left(c_{a,\uparrow}^{+}c_{b,\downarrow}^{+}-c_{a,\downarrow}^{+}c_{b,\uparrow}^{+}\right)^{2}$$





 $\frac{1}{2}\left(c_{a,\uparrow}^{+}c_{b,\downarrow}^{+}-c_{a,\downarrow}^{+}c_{b,\uparrow}^{+}\right)^{2}$

 $-\left(c_{a,\uparrow}^{+}c_{a,\downarrow}^{+}
ight)\left(c_{b,\uparrow}^{+}c_{b,\downarrow}^{+}
ight)$





$$\frac{\frac{1}{2}\left(c_{a,\uparrow}^{+}c_{b,\downarrow}^{+}-c_{a,\downarrow}^{+}c_{b,\uparrow}^{+}\right)^{2}}{=}$$

$$-\left(c_{a,\uparrow}^{+}c_{a,\downarrow}^{+}\right)\left(c_{b,\uparrow}^{+}c_{b,\downarrow}^{+}\right)$$
No preformed quartets
Glue between pairs
$$= \text{ interfaces with } R \sim \xi$$

$$- \text{ sign } \Rightarrow \pi \text{-junction}$$

$$I_{quartet} =$$

$$-|I_{c}| \sin(\varphi_{a} + \varphi_{b} - 2\varphi_{S})$$
Macroscopic
manifestation of the
internal structure of a
pair (orbital and spin
symmetries)

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Quartets in Metallic Structures A.H. Pfeffer, J.E. Duvauchelle, H. Courtois, R. Mélin, D. Feinberg and F. Lefloch, arXiv:1307.4862



Theoretical calculation

- Perturbative expansion in the tunnel amplitudes
- \Rightarrow Diffusion modes, evaluated in the ladder approximation

Quartets in Metallic Structures A.H. Pfeffer, J.E. Duvauchelle, H. Courtois, R. Mélin, D. Feinberg and F. Lefloch, arXiv:1307.4862



- Two-terminal SINIS ok with Usadel equations
- Quartet current if $eV > E_{th}$, not reduced as V increases

$$I_Q \propto E_{th} G_{CAR,NSN} \sin(\varphi_a + \varphi_b - 2\varphi_0)$$

Experimental Set-up A.H. Pfeffer, J.E. Duvauchelle, H. Courtois, and F. Lefloch



No Resonances for a Separated Bijunction A.H. Pfeffer, J.E. Duvauchelle, H. Courtois, R. Mélin, D. Feinberg, F. Lefloch, arXiv:1307.4862

2 resonances for direct Josephson $S_0 - S_a$ and $S_0 - S_b$



Resonances for a Bijunction (T = 200 mK) A.H. Pfeffer, J.E. Duvauchelle, H. Courtois, R. Mélin, D. Feinberg, F. Lefloch, arXiv:1307.4862



Resonances for a Bijunction A.H. Pfeffer, J.E. Duvauchelle, H. Courtois, R. Mélin, D. Feinberg, F. Lefloch, arXiv:1307.4862



Direct Josephson $S_a - S_0$ and $S_b - S_0$

Two resonances for $V_a = 0$ and $V_b = 0$

Resonances for a Bijunction A.H. Pfeffer, J.E. Duvauchelle, H. Courtois, R. Mélin, D. Feinberg, F. Lefloch, arXiv:1307.4862



Direct Josephson $S_a - S_b$

- Resonance expected at $V_a = V_b$
- However, not visible because lock-in excitation on S_0

Resonances for a Bijunction A.H. Pfeffer, J.E. Duvauchelle, H. Courtois, R. Mélin, D. Feinberg, F. Lefloch, arXiv:1307.4862



Three additional resonance lines

- $2V_0 = V_a + V_b$; $2V_a = V_0 + V_b$; $2V_b = V_0 + V_a$
- ullet Just permutation of the 3 terminals ightarrow equivalent resonances
- Are they due to quartets or to classical synchronization by an external impedance ?

Argument (A)

- Same external circuit for the bijunctions, separated or not
- No resonance for the separate bijunction
- ⇒ Resonance is due to the bijunction itself

Argument (B)

- Bijunction = phase-coherent mesoscopic object
- Nothing to do with classical resistance
- Usual RSJ descriptions of synchronization due non linear oscillators coupled by commmon shunt are not applicable

Argument (C)

- The strong resonant signal does not decay as *eV* is increased above *E*_{th}
 - AC Josephson oscillations decay above E_{th}
 - Quartets do not decay above E_{th}

Conclusion for the experimental part of the talk

 $(A)+(B)+(C) \Rightarrow$ Synchronization and resonances must be reconsidered from the begining, taking quartets into account.

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Generalization to Higher Order Resonances Exemple of the Sextet Resonance for $V_b = -2V_a$



Summary for the Quartet, Sextet Resonances



Summary for the Quartet, Sextet Resonances



Now let us put these resonances on a computer

The double Dot Structure T. Jonckheere, J. Rech, T. Martin, B. Douçot, D. Feinberg, R. Mélin, PRB '13



The starting Point of the Calculations T. Jonckheere, J. Rech, T. Martin, B. Douçot, D. Feinberg, R. Mélin, PRB '13



$$\begin{aligned} \mathcal{H}_{j} &= \sum_{k} \Psi_{jk}^{\dagger} \left(\xi_{k} \sigma_{z} + \Delta_{j} \sigma_{x} \right) \Psi_{jk} \\ \mathcal{H}_{D_{\alpha}} &= \epsilon_{\alpha} \sum_{\sigma = \uparrow, \downarrow} d_{\alpha\sigma}^{\dagger} d_{\alpha\sigma} \\ \hat{\mathcal{H}}_{T}(t) &= \sum_{jk\alpha} \Psi_{jk}^{\dagger} t_{j\alpha} e^{i\sigma_{z} \varphi_{j}/2} \mathbf{d}_{\alpha} + \text{h.c.} \end{aligned}$$

$$arphi_j(t) = arphi_j^{(0)} + 2 e V_j t / \hbar$$

$$\langle l_{j}(t)
angle = -2{\sf Re}\left\{{\sf tr}\left[\sigma_{z}\left(\hat{\Sigma}_{j}\otimes\hat{G}
ight)^{+,-}(t,t)
ight]
ight\}$$

Multipair Resonances in a double Dot Structure Metallic regime: T. Jonckheere, J. Rech, T. Martin, B. Douçot, D. Feinberg, R. Mélin, PRB '13



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In general, two Coexisting Transport Channels: Multipair Resonances and Phase-sensitive MARs T. Jonckheere, J. Rech, T. Martin, B. Douçot, D. Feinberg, R. Mélin, PRB '13





Quartets

With particle-hole symmetry:

Phase-MAR • Odd in voltage, Even in phase

- "cos($arphi_{a}+arphi_{b}-2arphi_{0}$) term", $I_{a}=-I_{b}$ (symmetric junction)
- Nonlocal quartets
 - Even in voltage, Odd in phase

• "sin(
$$\varphi_a + \varphi_b - 2\varphi_0$$
) term", $I_a = I_b$

Lowest-order Phase-MAR Process Triangular building block T. Jonckheere, J. Rech, T. Martin, B. Douçot, D. Feinberg, R. Mélin, PRB '13



What will be observed, Quartets, Multipairs or Phase-MAR ? Dot levels are at $(\epsilon_a, \epsilon_b) = (0.4\Delta, -0.4\Delta)$ T. Jonckheere, J. Rech, T. Martin, B. Douçot, D. Feinberg, R. Mélin, PRB '13

 π -shifted quasiequilibrium quartets

Giant resonance of nonequilibrium multipairs (in the sense of n + n pairs)

 $\mathsf{Phase}\mathsf{-}\mathsf{MAR}$



What will be observed, Multipairs or Phase-MAR ? T. Jonckheere, J. Rech, T. Martin, B. Douçot, D. Feinberg, R. Mélin, PRB '13



Total current is mainly

- In quartet or multipair channel at resonance
- In phase-MAR channel above the onset of phase-MARs

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BiSQUID J. Rech, T. Jonckheere, T. Martin, B. Douçot, D. Feinberg and R. Mélin, submitted to PRB



BiSQUID J. Rech, T. Jonckheere, T. Martin, B. Douçot, D. Feinberg and R. Mélin, submitted to PRB



BiSQUID: All 4 quantum dots at resonance J. Rech, T. Jonckheere, T. Martin, B. Douçot, D. Feinberg and R. Mélin, submitted to PRB



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Andreev interferometer (1 voltage, 1 phase) A.V. Galaktionov and A.D. Zaikin, PRB '13

3 superconducting contacts, one phase, one voltage



Lowest Order Phase-MAR Process (1/2) "Triangular" building block


A simplified Model for the Triangular Building Block (2/2) Next step in NSS' D. Gosselin, G. Hornecker, R. Mélin and D. Feinberg, PRB '14



- 1) \rightarrow 2): Electron propagation for NS_1 to S_1S_2 and Andreev reflection at S_1S_2
- 2) \rightarrow 3): CAR across S_1

The two Processes D. Gosselin, G. Hornecker, R. Mélin and D. Feinberg, PRB '14



Large transparency

Cooperative Andreev reflections

"Double Elastic Cotunneling" building block (dEC)



Small transparency

Pair/quasiparticle exchange

Triangular building block

Proposed Sample Configuration D. Gosselin, G. Hornecker, R. Mélin and D. Feinberg, PRB '14



Transparency-induced Cross-over in the Conductance D. Gosselin, G. Hornecker, R. Mélin and D. Feinberg, PRB '14



- Large transparency: spectroscopy in reflection
- Small transparency: spectroscopy in transmission

Typical Conductance Maps D. Gosselin, G. Hornecker, R. Mélin and D. Feinberg, PRB '14

- Phase and energy symmetry below the gap
- Phase-sensitive Tomash oscillations above the gap



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Provocative Conclusion: The End of the Subject ? Only five building blocks ! R. Mélin and D. Feinberg, PRB '04

This seminar concludes the investigation of all possible nonlocal transport electron-hole channels.

PHYSICAL REVIEW B 70, 174509 (2004)

Sign of the crossed conductances at a ferromagnet/superconductor/ferromagnet double interface

R. Mélin®

Centre de Recherches sur les Très Basses Températures (CRTBT⁺), CNRS, BP 166, 38042 Grenoble Cedex 9, France

D. Feinberg Laboratoire d'Eudes des Propriétés Electroniques des Solides (LEPES³), CNRS, BP 166, 38042 Grenoble Cedex 9, France (Received 12 July 2004; revised manuscript received 8 September 2004; published 16 November 2004)



- Elastic cotunneling (EC):
 - $\overline{g_{lpha,eta}^{1,1}g_{eta,lpha}^{1,1}}$ and $\overline{g_{lpha,eta}^{2,2}g_{eta,lpha}^{2,2}}$
- Cooper pair splitting (CAR):

$$\overline{g_{lpha,eta}^{1,2}g_{eta,lpha}^{2,1}}$$
 and $\overline{g_{lpha,eta}^{2,1}g_{eta,lpha}^{1,2}}$

• Double elastic cotunneling (dEC):

$$\overline{g_{lpha,eta}^{1,1}g_{eta,lpha}^{2,2}}$$
 and $\overline{g_{lpha,eta}^{2,2}g_{eta,lpha}^{1,1}}$

• Double crossed Andreev reflection

$$g^{1,2}_{lpha,eta}g^{1,2}_{eta,lpha}$$
 and $g^{2,1}_{lpha,eta}g^{2,1}_{eta,lpha}$

• Triangular or mixed:

$$g^{1,1}_{\alpha,\beta}g^{1,2}_{\beta,\alpha}$$
 and $g^{2,2}_{\alpha,\beta}g^{2,1}_{\beta,\alpha}$

Régis Mélin, Institut NEEL, Grenoble

Production of nonlocal quartets in a Josephson bijunction

Final Conclusion: Coming back to cross-correlations in NSN M. Flöser, D. Feinberg, R. Mélin, PRB '13 Expression of the cross-correlations, following Anantram and Datta (PRB '96)

$$\begin{split} S_{adk}(T = 0, V_{a}, V_{b}) \\ &= \frac{2e^{2}}{h} \int dE \\ &\times (2\pi[s_{ab}^{ab}s_{cb}^{ac}s_{bb}^{ad}] [\theta(|e|V_{a} - E) - 2\theta(|e|V_{a} - E)\theta(|e|V_{b} - E) + \theta(|e|V_{b} - E)] \\ &+ 2\pi[s_{ab}^{ab}s_{bb}^{ad}s_{bb}^{ad}] [\theta(|e|V_{a} - E) - 2\theta(|e|V_{a} - E)\theta(|e|V_{b} - E) + \theta(|e|V_{b} - E)] \\ &+ 2\pi[s_{ab}^{ab}s_{bb}^{ad}s_{bb}^{ad}] [\theta(|e|V_{a} - E) - 2\theta(|e|V_{a} - E)\theta(|e|V_{b} - E) + \theta(|e|V_{b} - E)] \\ &+ 2\pi[s_{ab}^{ab}s_{bb}^{ad}s_{bb}^{ad}s_{bb}^{ad}] [\theta(|e|V_{a} - E) + 2\theta(|e|V_{a} - E)\theta(|e|V_{b} - E) - \theta(|e|V_{b} - E)] \\ &+ 2\pi[s_{ab}^{ab}s_{bb}^{ad}s_{bb}^{ad}s_{bb}^{ad}] [-\theta(|e|V_{a} - E) + 2\theta(|e|V_{a} - E)\theta(|e|V_{b} - E) - \theta(|e|V_{b} - E)] \\ &+ 2\pi[s_{ab}^{ab}s_{bb}^{ad}s_{bb}^{ad}s_{bb}^{ad}] [-\theta(|e|V_{a} - E) + 2\theta(|e|V_{a} - E)\theta(|e|V_{b} - E) - \theta(|e|V_{b} - E)] \\ &+ 2\pi[s_{ab}^{ab}s_{bb}^{ad}s_{bb}^{ad}s_{bb}^{ad}] [-\theta(|e|V_{a} - E) + 2\theta(|e|V_{a} - E)\theta(|e|V_{b} - E) - \theta(|e|V_{b} - E)] \\ &+ 2\pi[s_{ab}^{ab}s_{bb}^{ad}s_{bb}^{ad}s_{bb}^{ad}] [\theta(|e|V_{a} - E) + 2\theta(|e|V_{a} - E)\theta(|e|V_{b} - E) - \theta(|e|V_{b} - E)] \\ &+ 2\pi[s_{ab}^{ab}s_{bb}^{ad}s_{bb}^$$

At perfect transparency and if $V_a = V_b$:

- All channels disappear except for EC-AR (pair fluctuations)
- Absence of CAR-NR (analogous of Cooper pair splitting for tunnel contacts)

Régis Mélin, Institut NEEL, Grenoble Production of nonlocal quartets in a Josephson bijunction

On-going Collaborations on Nonlocal Quartets in S_aSS_b NANOQUARTETS ANR project 2012-2015

NÉEL / INAC (Grenoble)

- Theory: Régis Mélin, Ciprian Padurariu, Denis Feinberg
- <u>Experiments</u>: Andreas Pfeffer, Jean-Eudes Duvauchelle, Hervé Courtois, François Lefloch

LPA-LPTHE (Paris)

- Theory: Audrey Cottet, Benoit Douçot
- Experiments: Laure Bruhat, Takis Kontos

CPT (Marseille)

• Theory: Thibaut Jonckheere, Jérôme Rech, Thierry Martin