

REPORT DOCUMENTATION PAGE

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RPPR Final Report

as of 25-May-2018

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Proposal Number: 71026ELII

Agreement Number: W911NF-17-1-0154

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Title: Size-selected nanocluster particles as novel building blocks for high-Tc superconducting systems

Begin Performance Period: 01-May-2017

End Performance Period: 28-Feb-2018

Report Term: 0-Other

Submitted By: Vitaly Kresin

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees:

STEM Participants: 3

Major Goals: The stated goal was “to formulate a roadmap and produce a testbed for the development of high-temperature superconducting electronic networks based on size-selected quantum-resolved nanocluster particles.”

The inspiration for this research came from two sources: (1) The USC group's experimental discovery in 2015 of superconducting pairing in individual aluminum nanocluster particles at approximately 120 K, which is about two orders of magnitude higher than in bulk aluminum (this effect should exist in other materials as well, and has the realistic potential to be extended up to room temperature); and (2) The fact that chains and networks built out of such nanoclusters soft-landed on a surface will be capable of carrying superconducting (and therefore lossless) currents due to tunneling between the nanoclusters (Josephson effect). Theory predicts that this current will be 100-1,000 times higher than in conventional Josephson systems, and operate at much higher temperatures.

Such a combination of size-selectively depositing nanoclusters, probing their individual superconductive properties, and working toward assembling them into organized superconducting networks had never been attempted previously. It requires state-of-the-art equipment for producing and mass-selecting bare metal nanoparticles, a means to soft-land them on a substrate of choice, imaging of the deposition result, and reliable tools for characterizing the transport properties.

The exploratory proposal was specifically focused on the task of optimizing the sample preparation and imaging steps, and on demonstrating that the samples can be accommodated on chips and mounts that are 100% usable for transport studies.

Specifically, the proposal's summary listed the following goals (abridged from the original for brevity):

- To employ the specialized deposition apparatus to soft-land size-selected metal nanocluster particles onto metallic single-wall carbon nanotubes, and to use Atomic Force Microscopy (AFM) followed by precision Transmission Electron Microscopy (TEM) to record and control the deposition process.
- To optimize the deposition steps by using surfaces decorated with single-walled carbon nanotubes (SWCNTs) from a solution, followed by deposition and imaging employing metallic SWCNT sensors. The latter are intended for subsequent templating and monitoring of high-Tc superconducting tunneling chains.
- To perform preliminary temperature-dependent conductivity measurements in a Physical Property Measurement System (PPMS) user facility

Accomplishments: To recap the aim stated in the original proposal, our goal was “to formulate a roadmap and produce a testbed for the development of high-temperature superconducting electronic networks based on size-selected quantum-resolved nanocluster particles.”

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The inspiration for this research came from two sources: (1) My group's experimental discovery in 2015 of superconducting pairing in individual aluminum nanocluster particles at approximately 120 K, which is about two orders of magnitude higher than in bulk aluminum (this effect should exist in other materials as well, and has the realistic potential to be extended up to room temperature); and (2) The fact that chains and networks built out of such nanoclusters soft-landed on a surface will be capable of carrying superconducting (and therefore lossless) currents due to tunneling between the nanoclusters (Josephson effect). Theory predicts that this current will be 100-1,000 times higher than in conventional Josephson systems, and operate at much higher temperatures.

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Our exploratory proposal was specifically focused on the task of optimizing the sample preparation and imaging steps, and on demonstrating that the samples can be accommodated on chips and mounts that are 100% usable for transport studies. Specifically, the proposal's summary listed the following goals (abridged from the original for brevity):

- To employ our specialized deposition apparatus to soft-land size-selected metal nanocluster particles onto metallic single-wall carbon nanotubes, and to use Atomic Force Microscopy (AFM) followed by precision Transmission Electron Microscopy (TEM) to record and control the deposition process.
- To optimize the deposition steps by using surfaces decorated with single-walled carbon nanotubes (SWCNTs) from a solution, followed by deposition and imaging employing metallic SWCNT sensors. The latter are intended for subsequent templating and monitoring of high-T_c superconducting tunneling chains.
- To perform preliminary temperature-dependent conductivity measurements in a Physical Property Measurement System (PPMS) user facility.

Accomplishments

The proposed goals have been met. Below is a brief summary of what has been accomplished.

1. A beam of nanocluster particles is produced by a magnetron sputtering/vapor condensation source followed by a mass filter and a deposition chamber. Since source behavior is controlled by a number of mutually interacting parameters, it is very important to understand this multi-variable optimization problem. We have carried out an extensive "factorial design" characterization of nanoparticle production. This work, has been published in the journal Applied Nanoscience under the title "Influence of source parameters on the growth of metal nanoparticles by sputter-gas-aggregation." The same paper describes the determination of nanocluster beam velocities, made with the help of a retarding-potential electrode arrangement which we designed and built. This data is essential for enabling low-energy ("soft") deposition of nanoclusters on substrates.

2. The central issue addressed in the exploratory proposal was to devise a means by which pairing in individual nanocluster particles could be detected, and which also could work as a template for bringing the nanoclusters together into prototype tunneling chains. The proposed solution was to use metallic SWCNT sensors which would be exposed to the nanocluster beam and "decorated" either with individual nanoparticles or with a "strand" of them. The very high sensitivity of nanotube sensors will make it possible to detect the onset of superconductivity in a nanocluster via the proximity effect, and the attachment of a series of nanoclusters along the tube will lead to the formation of network-like chains.

We have succeeded in our key goal, demonstrating that it is indeed possible to reliably position individual nanoparticles, as well as their sequences, along the wall of a metallic carbon nanotube sensor.

This work was presented at the 2018 APS March Meeting, and also at the 8th International Conference on the Science and Engineering of Novel Superconductors (Perugia, Italy) in July (invited talk) and at the American Vacuum Society Symposium (October 2018).

3. We were able to assemble and wire up the aforementioned SWCNT sensors within standard electronics chip

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mounts. These were used to run tests in a Quantum Design PPMS. The entire mount assembly (also a first for a nanoparticle deposition setup) has been tested for superconductivity with standard superconducting materials and verified to work perfectly.

The project now is ready to continue to observing a resistance transition in the nanotube sensors. Our test samples so far have been made with aluminum nanoparticles (because Al nanoclusters are the ones in which we have found a high-T_c transition), but in this form they cannot be used directly: aluminum oxidizes in air to a depth of several nanometers, hence sample transfer extinguishes the metallic character of most of the nanoparticle. We know exactly how to proceed: by building a “vacuum suitcase” for transfer to PPMS, and/or by mounting the deposition sample directly on a cryostat coldfinger for in situ measurements of T_c. Both are well-defined paths forward, and can be implemented in a straightforward manner within a continuation project. In addition, nanoparticles of other materials (e.g., tin) will be explored

4. We also have developed a method to deposit single-walled carbon nanotubes across TEM grids. We are performing size-selective deposition and electron microscope imaging on these samples. Importantly, the university’s electron microscopy center has acquired an energy-dispersive X-ray spectroscopy mapping detector which can provide chemical element sensitivity with ~2 angstrom resolution. This will allow us, for the first time, to determine the oxide thickness layer on metal nanoparticles as a function of size. This data will not only provide a major benchmark for understanding nanoscale oxidation, but it will be of immediate relevance to assessing tunneling junction thicknesses in nanoparticle and nanograin superconducting contacts.

In summary, the work performed under the auspices of this exploratory grant has validated the proposed nanosensor architecture for the development of superconducting circuits based on high-T_c size-resolved nanoclusters and nanocluster networks. Nanoparticle production has been characterized in detail, deposition on nanotube sensors demonstrated and its imaging accomplished, and versatile chip mounts designed. It is now viable to proceed to direct detection and demonstration of superconducting pairing and tunneling currents in these systems.

Training Opportunities: Two graduate students and one undergraduate have participated in the multiple facets of this project. They have learned a great deal about nanoparticle synthesis, mass spectrometry, deposition technology, nanofabrication tools, and imaging.

Results Dissemination: M. Khojasteh and V. V. Kresin, “Influence of source parameters on the growth of metal nanoparticles by sputter-gas-aggregation,” Applied Nanoscience 7, 875 (2017).

Poster abstract: American Physical Society March 2018 Meeting

Oral presentation abstract: American Vacuum Society Symposium (October 2018).

Invited talk abstract: 8th International Conference on the Science and Engineering of Novel Superconductors (Perugia, Italy, July 2018)

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: Vitaly Kresin

Person Months Worked: 1.00

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Funding Support:

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Participant Type: Graduate Student (research assistant)

Participant: Patrick Edwards

Person Months Worked: 9.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: Malak Khojasteh

Person Months Worked: 9.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Participant Type: Undergraduate Student

Participant: Diego Hernandez

Person Months Worked: 3.00

Funding Support:

Project Contribution:

International Collaboration:

International Travel:

National Academy Member: N

Other Collaborators:

Grant no. W911NF-17-1-0154, “Size-selected nanocluster particles as novel building blocks for high- T_c superconducting systems”

The aim stated in the original proposal was “to formulate a roadmap and produce a testbed for the development of high-temperature superconducting electronic networks based on size-selected quantum-resolved nanocluster particles.” The inspiration for this research came from two sources: (1) My group’s experimental discovery in 2015 of superconducting pairing in individual aluminum nanocluster particles at approximately 120 K, which is about two orders of magnitude higher than in bulk aluminum (this effect should exist in other materials as well, and has the realistic potential to be extended up to room temperature); and (2) The fact that chains and networks built out of such nanoclusters soft-landed on a surface will be capable of carrying superconducting (and therefore lossless) currents due to tunneling between the nanoclusters (Josephson effect). Theory predicts that this current will be 100-1,000 times higher than in conventional Josephson systems, and operate at much higher temperatures.

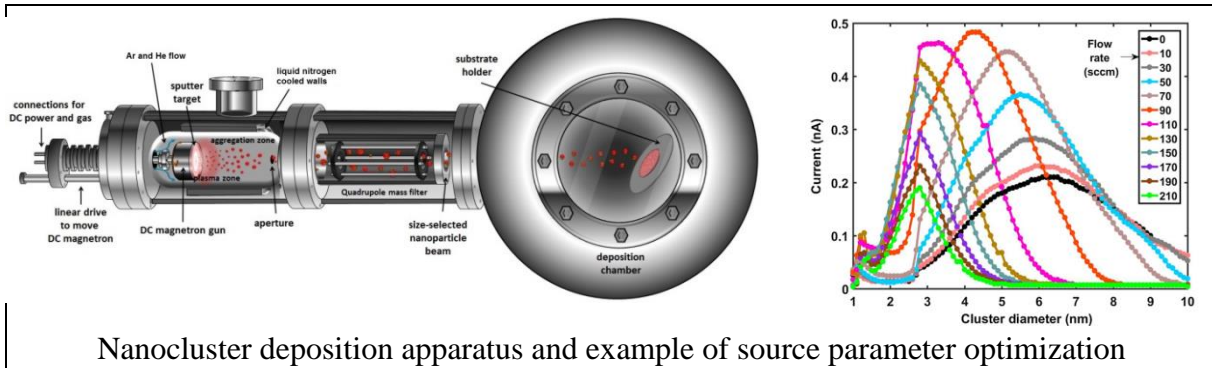
Such a combination of size-selectively depositing nanoclusters, probing their individual superconductive properties, and working toward assembling them into organized superconducting networks had never been attempted previously. It requires state-of-the art equipment for producing and mass-selecting bare metal nanoparticles, a means to soft-land them on a substrate of choice, imaging of the deposition result, and reliable tools for characterizing the transport properties.

Our exploratory proposal was specifically focused on the task of optimizing the sample preparation and imaging steps, and on demonstrating that the samples can be accommodated on chips and mounts that are 100% usable for transport studies. Specifically, the proposal’s summary listed the following goals (abridged from the original for brevity):

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- To perform preliminary temperature-dependent conductivity measurements in a Physical Property Measurement System (PPMS) user facility

I am pleased to report that these goals have been met. Below is a brief summary of what has been accomplished, accompanied by selected images.

1. A beam of nanocluster particles is produced by a magnetron sputtering/vapor condensation source followed by a mass filter and a deposition chamber, as illustrated in the figure below. Since source behavior is controlled by a number of mutually interacting parameters, it is very important to understand this multi-variable optimization problem. We have carried out an extensive “factorial design” characterization of nanoparticle production (see the figure).



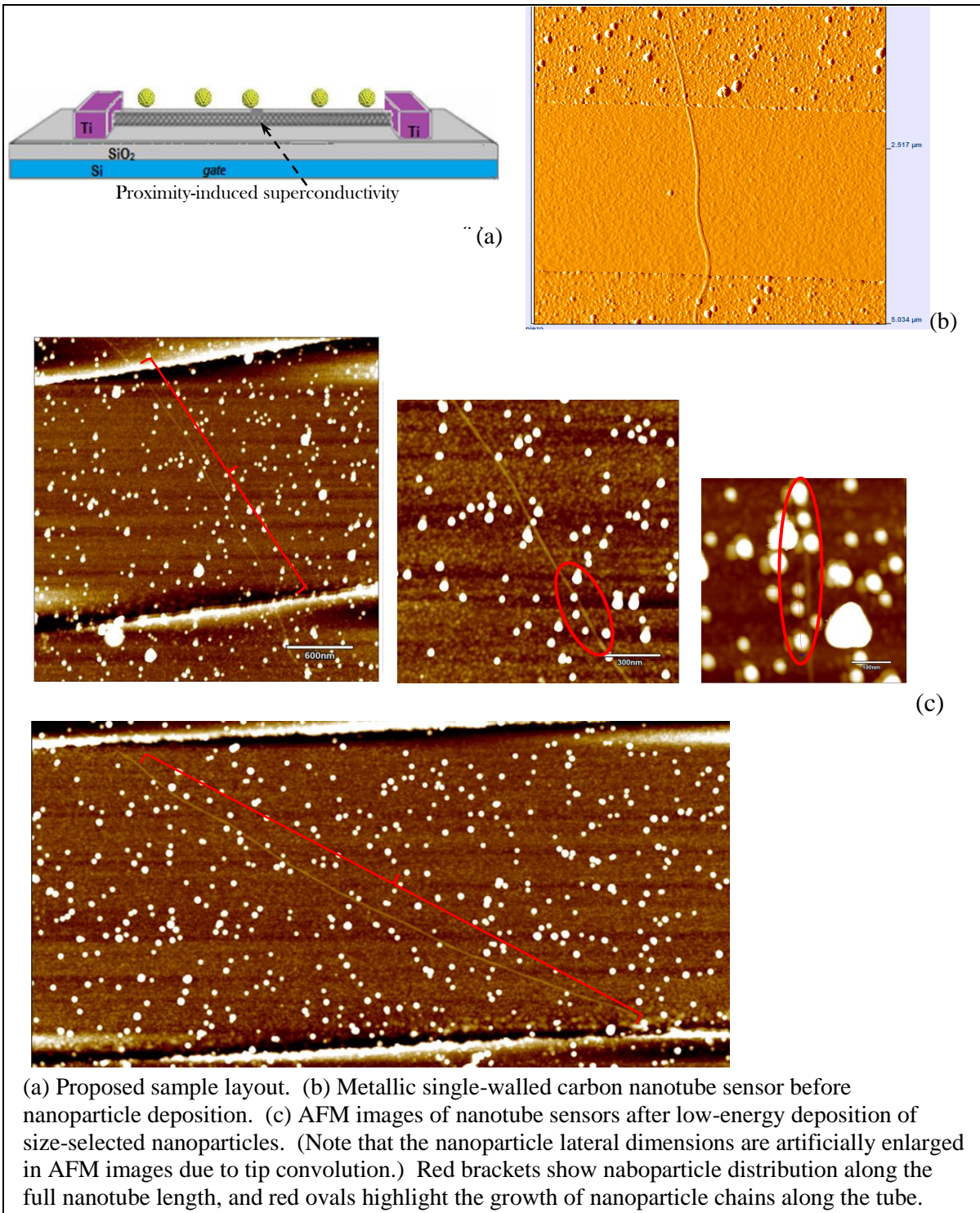
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The same paper describes the determination of nanocluster beam velocities, made with the help of a retarding-potential electrode arrangement which we designed and built. This data is essential for enabling low-energy (“soft”) deposition of nanoclusters on substrates.

2. The central issue addressed in the exploratory proposal was to devise a means by which pairing in individual nanocluster particles could be detected, and which also could work as a template for bringing the nanoclusters together into prototype tunneling chains. The proposed solution was to use metallic SWCNT sensors which would be exposed to the nanocluster beam and “decorated” either with individual nanoparticles or with a “strand” of them. The very high sensitivity of nanotube sensors will make it possible to detect the onset of superconductivity in a nanocluster via the proximity effect, and the attachment of a series of nanoclusters along the tube will lead to the formation of network-like chains.

The figure below shows the schematic outline from the original proposal, an image of a bare nanotube sensor, and images of the sensors after we performed nanoparticle deposition. It is seen that we have succeeded in our key goal: it is indeed possible to reliably position individual nanoparticles, as well as their sequences, along the wall of a metallic carbon nanotube sensor.

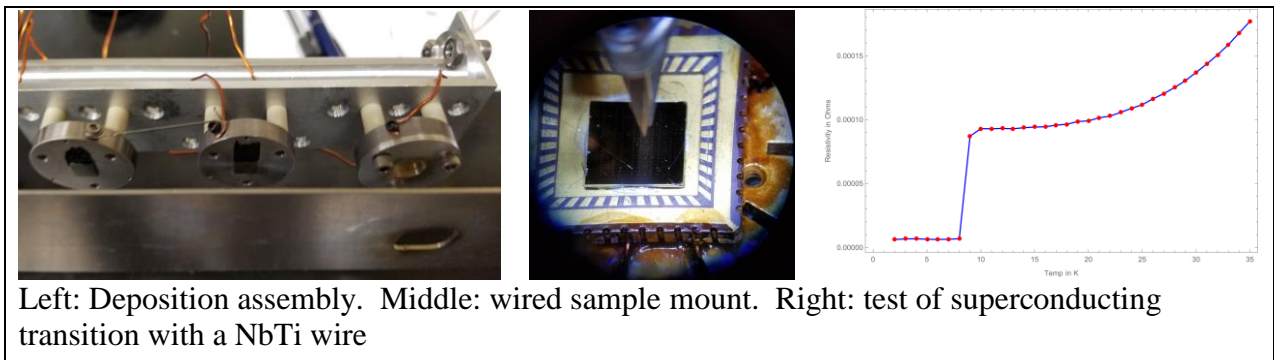
This work was presented at the 2018 APS March Meeting (poster), and will be presented also at the American Vacuum Society Symposium (oral contribution, October 2018) and at the 8th International Conference on the Science and Engineering of Novel Superconductors (Perugia, Italy) in July (invited talk).



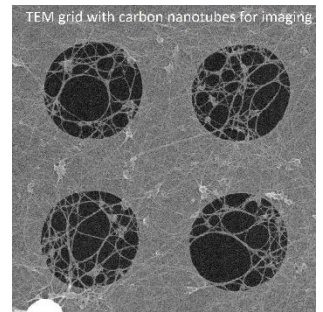
3. We were able to assemble and wire up the aforementioned SWCNT sensors within standard electronics chip mounts. These were used to run tests in a Quantum Design PPMS. The entire mount assembly (also a first for a nanoparticle deposition setup) has been tested for

superconductivity with a sample of a thin NbTi wire, and shown to work perfectly (see the figure below).

The project now is ready to continue to observing a resistance transition in the nanotube sensors. Our test samples so far have been made with aluminum nanoparticles (because Al nanoclusters are the ones in which we have found a high- T_c transition), but in this form they cannot be used directly: aluminum oxidizes in air to a depth of several nanometers, hence sample transfer extinguishes the metallic character of most of the nanoparticle. We know exactly how to proceed: by building a “vacuum suitcase” for transfer to PPMS, and/or by mounting the deposition sample directly on a cryostat coldfinger for *in situ* measurements of T_c . Both are well-defined paths forward, and can be implemented in a straightforward manner in a continuation project. In addition, nanoparticles of other materials (e.g., tin) will be explored.



3. We also have developed a method to deposit single-walled carbon nanotubes across TEM grids. We are performing size-selective deposition on these samples, and within several weeks expect to have detailed electron microscope images of individual nanoparticles. Importantly, the university’s electron microscopy center has acquired an energy-dispersive X-ray spectroscopy mapping detector which can provide chemical element sensitivity with $\sim 2 \text{ \AA}$ resolution. This will allow us, for the first time, to determine the oxide thickness layer on metal nanoparticles as a function of size. This data will not only provide a major benchmark for understanding nanoscale oxidation, but it will be of immediate relevance to assessing tunneling junction thicknesses in nanoparticle and nanograin superconducting contacts.



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