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RPPR Final Report
as of 25-Mar-2020

Agency Code:

Proposal Number: 71740PHDRP **Agreement Number: W911NF-17-1-0348**
INVESTIGATOR(S):

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Report Date: 31-Aug-2019 Date Received: 29-Aug-2019
Final Report for Period Beginning 01-Sep-2017 and Ending 31-May-2019
Title: Metamaterial Superconductors
Begin Performance Period: 01-Sep-2017 **End Performance Period:** 31-May-2019
Report Term: 0-Other
Submitted By: Vera Smolyaninova Email: vsmolyaninova@towson.edu
Phone: (410) 704-2608
Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 4 **STEM Participants:** 6

Major Goals: The goal is to apply metamaterial approaches of effective dielectric response function engineering to niobium titanium nitride (NbTiN) and magnesium diboride (MgB2) for improvement of their superconducting properties. Unlike traditional searches for higher Tc superconductors among natural materials, which exhibit larger electron-electron interactions by virtue of chance, we suggest to engineer the electron pairing interaction using the unique assortment of tools, which have been very recently developed in the field of artificial electromagnetic metamaterials. Considerable enhancement of attractive electron-electron interaction may be expected hyperbolic metamaterials (multilayered superconductor/dielectric heterostructures). Such NbTiN and MgB2 need to be fabricated and their dielectric function and superconducting properties need to be tested and correlated.

Accomplishments: Please see attached file.

Training Opportunities: Towson University

Undergraduate students Sabrina Searfoss, Bryan Augstein, and William Korzi worked on this project. They have learned different optical characterization techniques such as FTIR, polarization reflectometry and were exposed to ellipsometry and low temperature transport measurements. Sabrina and Will were trained to fit polarization reflectometry data for determination of optical constants. These students participated in the Towson Research & Creative Inquiry Forum, Spring 2018 and 2019. William Korzi presented his work on this project at the March Meeting of the American Physical Society, Boston MA, 2019. Bryan Augstein graduated in May 2018 with B.Sc. in Physics and was admitted to the Professional Masters Program in Applied Physics at Towson. He graduated with Masters Degree in Applied Physics in August 2019.

Graduate masters student William Zimmerman participated in project-related activities through taking research courses for academic credit. He have learned FTIR, polarization reflectometry and low temperature transport measurements techniques. William participated in MAS-APS Meeting 2017 and in the Towson Research & Creative Inquiry Forum, Spring 2018. William Zimmerman graduated in May 2018 with Masters Degree in Applied Physics.

A post-doctoral researcher Dr. Grace Yong have learned different optical characterization techniques such as FTIR, polarization reflectometry and ellipsometry measurements. She developed a fitting routing for determination of optical constants from polarization reflectometry data.

The PI participated in SPIE 2017 and 2019, META2018, APS March Meeting 2019 conferences.

Temple University

PhD graduate students Narendra Acharya, Wenura K. Withanage, Kanishka Wijesekara, and Fei Qin worked on developing metamaterial multilayers.

RPPR Final Report
as of 25-Mar-2020

Results Dissemination: Peer-reviewed publication:

1. I. I. Smolyaninov and V. N. Smolyaninova, “Metamaterial superconductors”, Nanophotonics 7, 795–818, (2018)
2. Igor I. Smolyaninov, Vera N. Smolyaninova, “Enhancement of Coulomb blockade in epsilon near zero and hyperbolic metamaterials,” Physica C 556, 14 (2019)
3. Vera N. Smolyaninova, William Korzi, William Zimmerman, Sabrina Searfoss, Christopher Jensen, Grace Yong, David Schaefer, Joseph C. Prestigiacomo, M.S. Osofsky, Kim Heungsoo, Xing Zhen, M.M. Qazilbash, Igor I. Smolyaninov, “Superconducting properties of tin-based ENZ and hyperbolic metamaterials,” Physica C 556 (2019) 14–18
4. Vera N. Smolyaninova, Jeffrey W. Lynn, Nicholas P. Butch, Heather Chen-Mayer, Joseph C. Prestigiacomo, M. S. Osofsky, and Igor I. Smolyaninov, “Observation of plasmon-phonons in a metamaterial superconductor using inelastic neutron scattering,” PHYSICAL REVIEW B 100, 024515 (2019)

Conference presentations:

1. Osofsky M., Smolyaninova V., Zander K., Gresock T., Saha S., Yost B., Jensen C., Prestigiacomo J., Kim H., Bassim N., Greene R., Smolyaninov I., “Application of metamaterial nano-engineering for increasing the superconducting critical temperature,” 28th International Conference on Low Temperature Physics, Gothenburg, Sweden, 2017
2. Vera N. Smolyaninova, Christopher Jensen, William Zimmerman, Joseph C. Prestigiacomo, Michael S. Osofsky, Heungsoo Kim, Nabil D. Bassim, Zhen Xing, Mumtaz Qazilbash, Igor I. Smolyaninov, “Enhanced superconductivity in hyperbolic metamaterials” (invited), SPIE 2017, San Diego, CA
3. Osofsky M., Smolyaninova V., Zander K., Gresock T., Saha S., Yost B., Jensen C., Prestigiacomo J., Kim H., Bassim N., Greene R., Smolyaninov I., “Application of metamaterial nano-engineering for increasing the superconducting critical temperature,” 13th European Conference on Applied Superconductivity, Geneva, Switzerland, 2017
4. William Zimmerman, Sabrina Searfoss, Christopher Jensen, Grace Yong, Vera Smolyaninova, Joseph Prestigiacomo, M. S. Osofsky, Igor Smolyaninov, “Enhanced superconductivity in tin-based hyperbolic metamaterials,” MAS-APS Meeting 2017, Newark, New Jersey
5. Michael S. Osofsky, Vera N. Smolyaninova, Kathryn Zander, Thomas Gresock, Shanta Saha, Bradley Yost, Christopher Jensen, Joseph Prestigiacomo, Heungsoo Kim, Nabil D. Bassim, Richard Greene and Igor Smolyaninov, “Application of Metamaterial Nanoengineering for Increasing of Superconducting Critical Temperature,” MRS Fall Meeting, Boston, MA 2017
6. Osofsky M., Smolyaninova V., Zander K., Gresock T., Saha S., Yost B., Jensen C., Prestigiacomo J., Kim H., Bassim N., Greene R., Smolyaninov I., “Application of metamaterial nano-engineering for increasing the superconducting critical temperature” (invited), EMN Orlando Meeting 2017, Orlando FA
7. Michael Osofsky, Vera Smolyaninova, Joseph Prestigiacomo and Igor Smolyaninov, “Application of metamaterial nano-engineering for increasing the superconducting critical temperature,” CMCEE 2018, Singapore.
8. Vera Smolyaninova and Igor Smolyaninov, “Metamaterial superconductors” (invited), META 2018 Conference, the 9th International Conference on Metamaterials, Photonic Crystals and Plasmonics, Marseille, France.
9. Michael Osofsky, Vera Smolyaninova, Joseph Prestigiacomo, Heungsoo Kim, Nabil Bassim, Richard Greene, Xiaoxing Xi, Igor Smolyaninov, “Application of metamaterial nano-engineering for increasing the superconducting critical temperature,” Applied Superconductivity Conference, Seattle, Washington, 2018
10. Vera Smolyaninova, Jeffrey W. Lynn, Nicholas Butch, Heather Chen-Mayer, Joseph Prestigiacomo, Michael Osofsky, Igor Smolyaninov, “Observation of plasmon-phonons in a metamaterial superconductor using inelastic neutron scattering,” APS March Meeting, Boston MA, 2019
11. Will Korzi, Grace Yong, Bryan Augstein, Wenura K Withanage, Fei Qin, Kanishka Wijesekara, Narendra Acharya, Xiaoxing Xi, Anne-Marie Valente-Feliciano, Joseph Prestigiacomo, Michael Osofsky, Igor Smolyaninov, Vera Smolyaninova, “MgB2 and NbTiN-based hyperbolic metamaterial superconductors,” APS March Meeting, Boston MA, 2019
12. Michael Osofsky, Vera Smolyaninova, Joseph Prestigiacomo, Peter Rosen, Matthew Dickson, Brian Woodfield, Jeffrey W. Lynn, Nicholas Butch, Heather Chen-Mayer, Igor Smolyaninov, “Application of metamaterial nano-engineering for increasing the superconducting critical temperature,” APS March Meeting, Boston MA, 2019

Outreach:

Conducted a lab/research tour for STEM female students from Richmond Hall Towson University.

RPPR Final Report
as of 25-Mar-2020

Honors and Awards: Vera Smolyaninova was awarded 2018 University System of Maryland Board of Regents Faculty Award for Excellence in Scholarship/Research

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI
Participant: Vera Smolyaninova
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co PD/PI
Participant: Anne-Marie Valente-Feliciano
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co PD/PI
Participant: Michael Osofsky
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co PD/PI
Participant: Xiaoxing Xi
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co-Investigator
Participant: Joseph Prestigiacomo
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

RPPR Final Report
as of 25-Mar-2020

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Grace Yong
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Graduate Student (research assistant)
Participant: Fei Qin
Person Months Worked: 3.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Graduate Student (research assistant)
Participant: Wenura Withanage
Person Months Worked: 8.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Graduate Student (research assistant)
Participant: Bryan Augstein
Person Months Worked: 4.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Undergraduate Student
Participant: Bryan Augstein
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Undergraduate Student
Participant: William Korzi
Person Months Worked: 6.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

RPPR Final Report
as of 25-Mar-2020

Participant Type: Undergraduate Student
Participant: Sabrina Searfoss
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

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Date Submitted: 10/7/18 12:00AM Date Published: 3/8/18 10:00AM
Publication Location: Germany
Article Title: Metamaterial superconductors
Authors: I.I. Smolyaninov, V.N. Smolyaninova
Keywords: superconductivity; electromagnetic metamaterials; dielectric properties; epsilon near-zero metamaterial; hyperbolic metamaterial
Abstract: Searching for natural materials exhibiting larger electron-electron interactions constitutes a traditional approach to high-temperature superconductivity research. Very recently, we pointed out that the newly developed field of electromagnetic metamaterials deals with the somewhat related task of dielectric response engineering on a sub-100-nm scale. Considerable enhancement of the electron-electron interaction may be expected in such metamaterial scenarios as in epsilon near-zero (ENZ) and hyperbolic metamaterials. In both cases, dielectric function may become small and negative in substantial portions of the relevant four-momentum space, leading to enhancement of the electron pairing interaction. This approach has been verified in experiments with aluminum-based metamaterials. Metamaterial superconductor with Tc=3.9 K have been fabricated, which is three times that of pure aluminum (Tc=1.2 K), which opens up new possibilities to improve the Tc of other simple superconductors consider
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Volume: 556 Issue: First Page #: 14
Date Submitted: 8/26/19 12:00AM Date Published: 11/7/18 5:00AM
Publication Location:
Article Title: Enhancement of Coulomb blockade in epsilon near zero and hyperbolic metamaterials
Authors: Igor I. Smolyaninov, Vera N. Smolyaninova
Keywords: Metamaterial Superconductor Coulomb blockade
Abstract: Coulomb interaction of charges in a metamaterial may be expressed via its effective dielectric response function. Coulomb interaction appears considerably enhanced in artificially engineered epsilon near zero (ENZ) and hyperbolic metamaterials. Indeed, very recently it was observed that negative ENZ conditions enhance superconducting properties of a composite metamaterial via enhancement of Cooper pairing. Here we demonstrate that positive ENZ conditions enhance Coulomb repulsion of charges, leading to considerable enhancement of the Coulomb blockade effect in such metamaterials. Similar to high Tc cuprates, in hyperbolic metamaterials superconductivity and the Coulomb blockade-induced charge ordering is predicted to compete with each other.
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RPPR Final Report
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Volume: 565 Issue: First Page #: 1353511
Date Submitted: 8/26/19 12:00AM Date Published: 7/29/19 4:00AM
Publication Location:
Article Title: Superconducting properties of tin-based ENZ and hyperbolic metamaterials
Authors: 3. Vera N. Smolyaninova, William Korzi, William Zimmerman, Sabrina Searfoss, Christopher Jensen,
Keywords: Superconductivity Metamaterial Tin
Abstract: Recent experiments have demonstrated that the superconducting critical temperature may be improved in various metamaterial superconductor geometries. Here, we present the results of a study of tin-based metamaterial superconductors in the epsilon-near-zero (ENZ) and hyperbolic metamaterial configurations. It was observed that Tc enhancement is significantly reduced when the metamaterial structural dimensions exceed 240 nm, the superconducting coherence length in pure tin.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
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Volume: 100 Issue: 2 First Page #: 024515
Date Submitted: 8/26/19 12:00AM Date Published: 7/1/19 4:00AM
Publication Location:
Article Title: Observation of plasmon-phonons in a metamaterial superconductor using inelastic neutron scattering
Authors: Vera N. Smolyaninova, Jeffrey W. Lynn, Nicholas P. Butch, Heather Chen-Mayer, Joseph C. Prestigiacc
Keywords: superconductivity, metamaterials, plasmon-phonon
Abstract: metamaterial approach is capable of drastically increasing the critical temperature, Tc, of composite metal-dielectric superconductors as demonstrated by the tripling of Tc that was observed in bulk Al-Al2O3 core-shell metamaterials. A theoretical model based on the Maxwell-Garnett approximation provides a microscopic explanation of this effect in terms of electron-electron pairing mediated by a hybrid plasmon-phonon excitation. We report an observation of this excitation in Al-Al2O3 core-shell metamaterials using inelastic neutron scattering. This result provides support for this mechanism of superconductivity in metamaterials.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

CONFERENCE PAPERS:

Publication Type: Conference Paper or Presentation **Publication Status:** 0-Other
Conference Name: 28th International Conference on Low Temperature Physics
Date Received: Conference Date: 15-Oct-2018 Date Published: 15-Oct-2018
Conference Location: Gothenburg, Sweden
Paper Title: Application of metamaterial nano-engineering for increasing the superconducting critical temperature
Authors: Osofsky M., Smolyaninova V., Zander K., Gresock T., Saha S., Yost B., Jensen C., Prestigiacomo J., Ki
Acknowledged Federal Support: **Y**

RPPR Final Report
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Conference Name: SPIE 2017
Date Received: Conference Date: Date Published:
Conference Location: San Diego CA
Paper Title: Enhanced superconductivity in hyperbolic metamaterials
Authors: Vera N. Smolyaninova, Christopher Jensen, William Zimmerman, Joseph C. Prestigiacomo, Michael S.
Acknowledged Federal Support: **Y**

Accomplishments

MgB_2-based multilayers

Three approaches were used to grow MgB_2/dielectric multilayers.

Approach 3: Direct deposition of ultra-thin MgB_2 with MgO as a dielectric.

5 nm MgB_2 films were deposited directly on SiC and MgO using direct hybrid physical chemical vapor deposition (HPCVD) growth of ultra-thin film and MgO sputtering. MgB_2/MgO multilayers were prepared by repeating these steps. Single layer MgB_2 and 4-layer multilayers with different MgO thicknesses were grown.

Heating of the first layer in preparation for the deposition of the second layer leads to the decrease of the T_c of this layer (Fig. 1). **In the 4-layers the T_c is restored, or slightly increased (probably due to metamaterial effect)**, but does not exceed the T_c of not-thermally treated single layer. The resistivity of multilayers is more than one order of magnitude higher than that of the single layers of similar thicknesses.

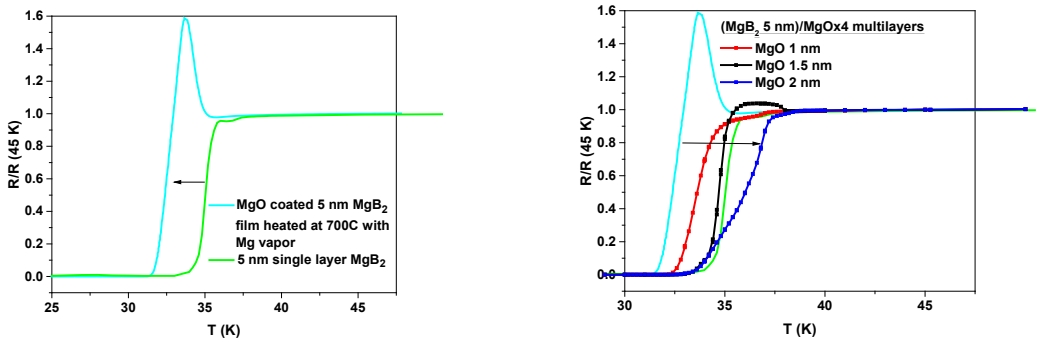

Fig. 1. Temperature dependence of normalized resistance of (5 nm MgB_2)/MgO multilayers with different thicknesses of MgO.

Fig. 2 shows reflectivity of the samples measured with Fourier Transform Infra-Red spectroscopy (FTIR). Thin single layer samples and x4 multilayer samples have low reflectivity

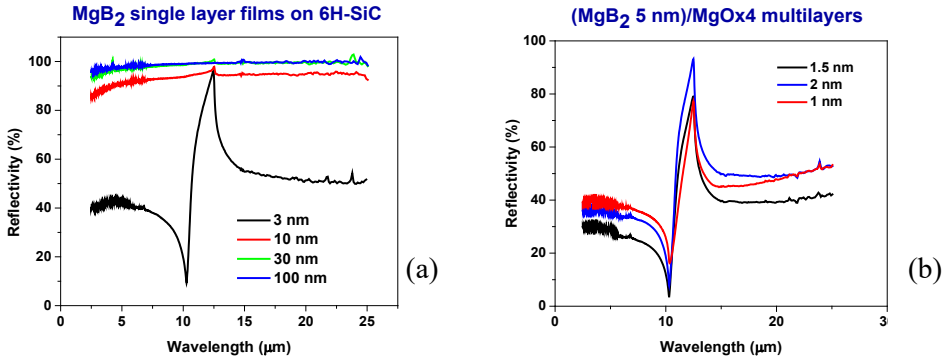

Fig. 2. Reflectivity of single layer of MgB_2 of different thicknesses (a) and (5 nm MgB_2)/MgO multilayers with different thicknesses of MgO measured with FTIR (b).

and show properties of the substrate (phohon-polariton resonance (PPR) in SiC at around 11 micron). The thicker films (starting from 10 nm) appear much more metallic and do not exhibit this behavior. The reflectivity of multilayers of total thickness of MgB_2 of 20 nm should be sufficiently high. However, **the reflectivity of (5 nm MgB_2)/MgO samples (Fig. 2) is too to exhibit hyperbolic characteristics.** Such low reflectivity is consistent with low conductivity values.

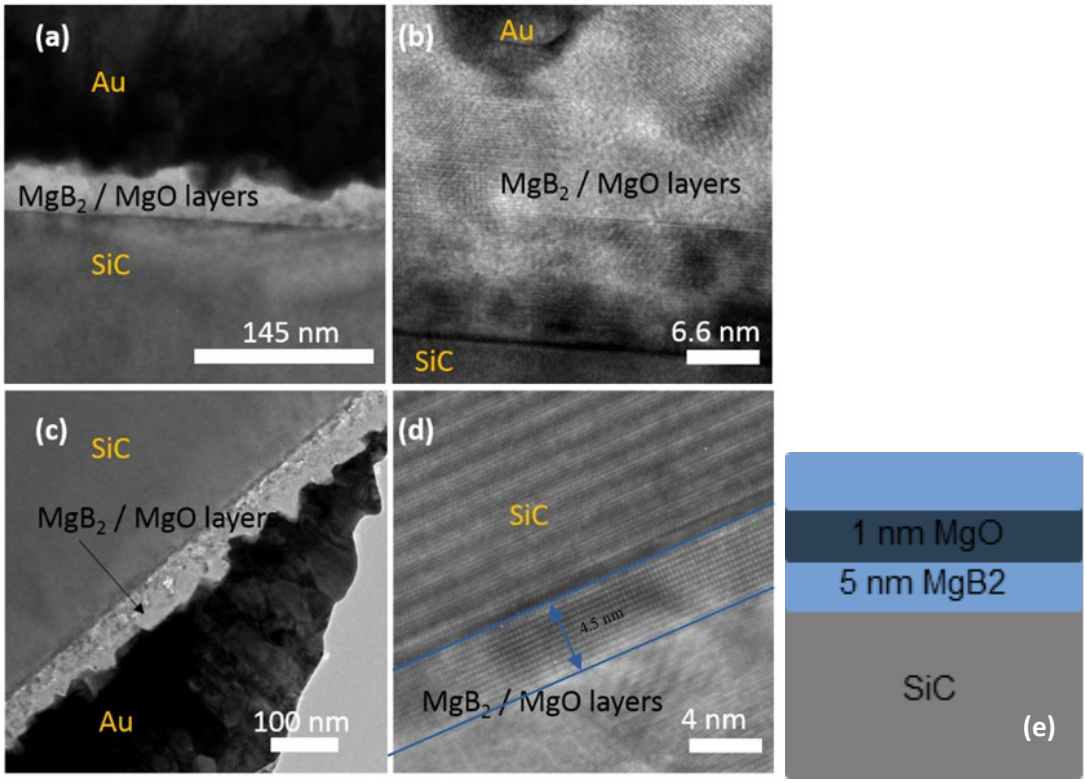

Fig. 3. Transmission Electron Microscopy (TEM) images: (a) and (b): (1.5 nm MgO/5 nm MgB_2) x 4 (c) and (d): (2 nm MgO/5 nm MgB2) x 4; (e) schematic sequence of layers.

TEM images (Fig. 3) showed that no clear layer structure were observed. Only the first 5 nm MgB_2 layer was clearly visible. **The surface roughness of each successive layer becomes progressively higher, which prevents forming well defined subsequent layers.**

Approach 2: Use AlN for dielectric layer.

Single layer MgB_2 and 8-layer multilayers with different AlN thicknesses were grown. Multilayers with 2 nm of AlN have smaller T_c than single layer of MgB_2/AlN. Multilayers with 1 nm of AlN have slightly larger T_c than single layer of MgB_2/AlN, but not larger than a single layer MgB_2 of similar thickness. Resistivity of MgB_2/AlN multilayers is at least one order of magnitude higher than the resistivity of 10 nm single layer MgB_2 film. These multilayers do not exhibit hyperbolic metamaterial properties because of low conductivity of the layers. The surface roughness of each successive layer accumulates up to 15 nm. **It is likely that surface roughness**

leads to decline of the conductive properties of the multilayers. The surface roughness could be improved with the ion milling of each layers.

Approach 1: Ar ion milling of thick MgB_2 films to produce ultra-thin MgB_2 layer with MgO or AlN as the dielectric layer.

To improve the surface roughness the following fabrication steps were used:

- HPCVD of thick (~ 15 nm) MgB_2 film on SiC
- Ar ion milling at low angle to reduce the thickness the MgB_2 film to achieve ultra-thin MgB_2 films
- In-situ sputtering of MgO ultra-thin layer
- HPCVD deposition of 2nd thick MgB_2 layer
- Ar ion milling at low angle to reduce the thickness the MgB_2 film to achieve ultra-thin MgB_2 films
- In-situ sputtering of MgO ultra-thin layer
- Continue this process to achieve multilayers

This method did not help to prevent the following issues:

- Polycrystalline growth of 2^{nd} MgB_2;
- Increased roughness of the 2^{nd} layer;
- Increased roughness after ion milling of 2nd MgB_2 layer (roughness is close to the expected thickness).

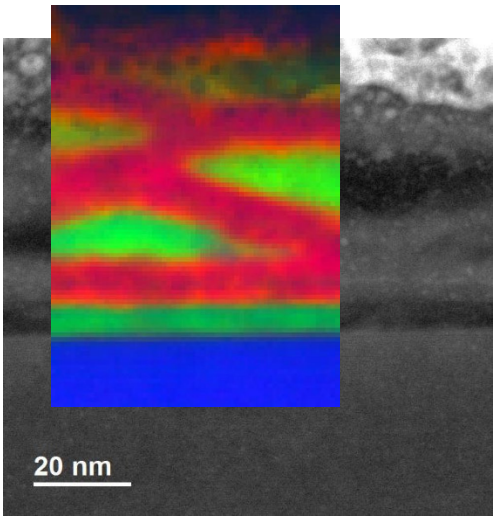

Fig. 4. TEM image of MgB_2/MgO multilayer. Color map based on linear least-squares combination of characteristic EELS plasmon shapes for each material: MgO - red; MgB_2 – green; Si – blue.

The TEM image of this sample (Fig. 4) showed that only the first layer is grown epitaxially, but **the subsequent layers are intermixed**.

The temperature dependence of resistance of these samples is shown in Fig. 5. Imperfections in layers and inhomogeneity leads to variations in the T_c in nominally the same samples and to the broadness of the transition. The resistivity of multilayers is more than one order of magnitude higher than that of the single layers of similar thicknesses.

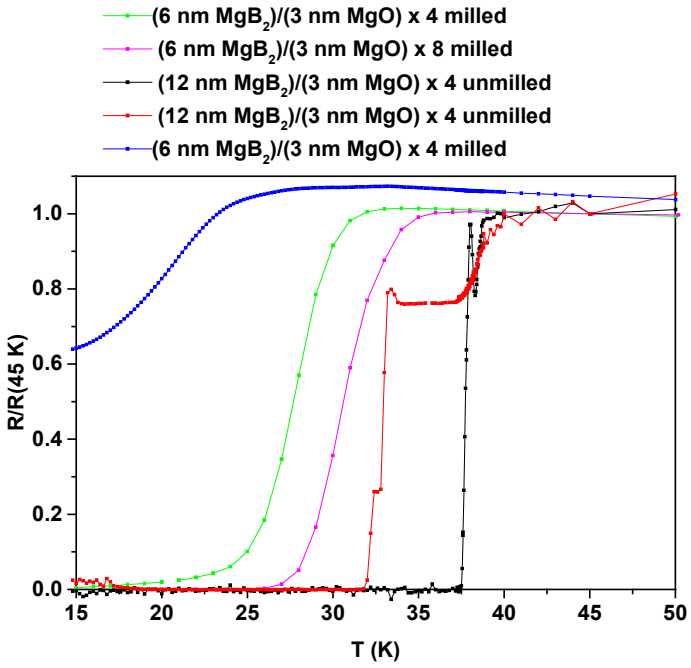

Fig. 5. Temperature dependence of normalized resistance of MgB_2/(3 nm MgO) multilayers with milled and unmilled MgB_2 layers.

Characterization of the electromagnetic properties of the fabricated multilayers was done using polarization reflectometry techniques. Polarization reflectometry was used to determine the signs of the real part of in-pane (ε_1) and out of plane (ε_2) dielectric functions, and thus the hyperbolic metamaterial character of the fabricated samples. Reflectivity for s-polarization is given in terms of the incident angle θ by

$$R_s = \left| \frac{\sin(\theta - \theta_{ts})}{\sin(\theta + \theta_{ts})} \right|^2 , \quad (1)$$

where

$$\theta_{ts} = \arcsin\left(\frac{\sin\theta}{\sqrt{\varepsilon_1}} \right) . \quad (2)$$

Reflectivity for p-polarization is given as

$$R_p = \left| \frac{\varepsilon_1 \tan\theta_{tp} - \tan\theta}{\varepsilon_1 \tan\theta_{tp} + \tan\theta} \right|^2 , \quad (3)$$

where

$$\theta_{tp} = \arctan\sqrt{\frac{\varepsilon_2 \sin^2\theta}{\varepsilon_1\varepsilon_2 - \varepsilon_1 \sin^2\theta}}. \qquad (4)$$

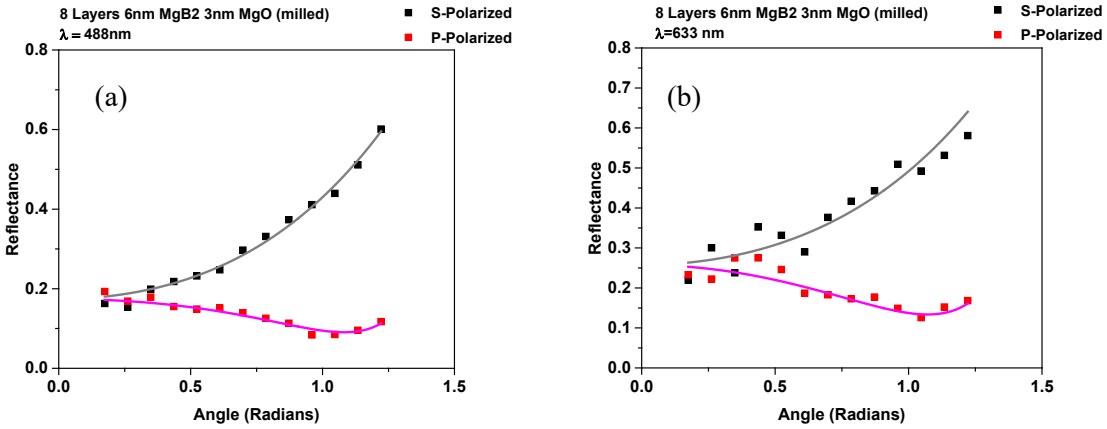

Fig. 6. Polarization reflectometry data for 8 layers (6 nm MgB_2)/(3 nm MgO) multilayers measured for λ = 488 nm (a) λ = 633 nm (b). Solid lines are fits to Eqs. (1 – 4).

Polarization reflectometry data for 8 layers (6 nm MgB_2)/(3 nm MgO) multilayers are show in Fig. 6. Data were fitted with Eqs. (1 – 4). These fits determined the following dielectric functions:

λ = 488 nm

$\varepsilon_{1\ (in\ plane)}$= 0.098 + 1.7i
$\varepsilon_{2\ (out\ of\ plane)}$= 1.3 + 0.5i

λ = 633 nm

$\varepsilon_{1(in\ plane)}$= 0.4 + 4.1i

$\varepsilon_{2(out\ of\ plane)}$= 5 + 6.2i

(6 nm MgB_2/3 nm MgO)x8 multilayers are not hyperbolic in the visible range: $\varepsilon_{in\ plane}$ and $\varepsilon_{out\ of\ plane}$ are positive because of i**nsufficient conductivity and imperfections of MgB_2 layers**

In another attempt to improve surface roughness, the 6 nm MgB_2 was made by growing 20 nm MgB_2 first and then thinned down to 6 nm using ion milling. Larger than optimal thickness of MgB_2 was chosen to help improve the surface roughness. The 3 nm of amorphous AlN was grown by DC magnetron sputtering at temperature of 700 C, since higher temperature will damage the superconductivity of MgB_2 thin film. 8 layers multilayer of (6 nm MgB_2)/(AlN) was fabricated. The FTIR reflectivity of the sample has increased, and the resistivity of the sample has become similar to resistivity of 6 nm thin single layer, showing that conducting properties of MgB_2 layers were not damaged during fabrication.

Polarization reflectometry measurements showed that 8 layers multilayer of (6 nm MgB_2)/(3 nm AlN) are not hyperbolic in the visible region, but exhibit hyperbolic behavior for λ

= 1550 nm (Fig. 7). However, the T_c of this sample remain similar to the T_c of single 6 nm layer of MgB_2.

For metamaterial approach to be successful, **the dimension of the metamaterial unit (MgB_2/dielectric bilayer) should be smaller than coherence length**. However, the thickness of bilayer, 9 nm, exceeds the coherence length of MgB_2. Unfortunately, **a good quality thinner layers was not possible to fabricate using described methods.**

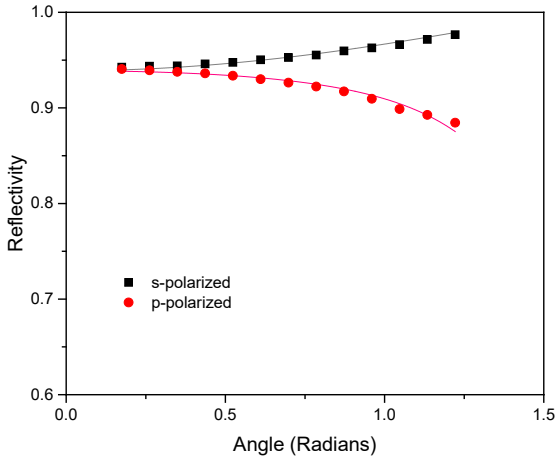

Fig. 7. Polarization reflectometry data for 8 layers (6 nm MgB_2)/(3 nm AlN) multilayers measured for λ = 1550 nm. Solid lines are fits to Eqs. (1 – 4) with parameters:

$\varepsilon_{1\ (in\ plane)} = -36.2 + 28.8i$

$\varepsilon_{2\ (out\ of\ plane)} = 16 + 1.5i$

NbTiN- based multilayers

NbTiN/AlN multilayers were deposited using reactive high power impulse magnetron sputtering (R-HiPIMS). (3 nm NbTiN)/AlN multilayers with 1, 2, 4, 8 and 16 layers and AlN thickness of 1, 1.5 and 2 nm were grown on MgO substrate. The TEM image of (3 nm NbTiN)/(2 nm AlN) x 17 multilayer sample (Fig. 8) shows well-defined layers.

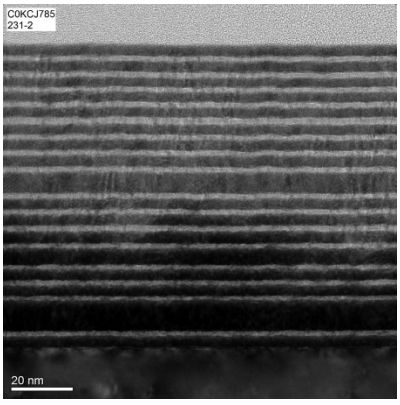

Fig. 8. TEM images of (3 nm NbTiN)/(2 nm AlN) x 16 multilayer sample.

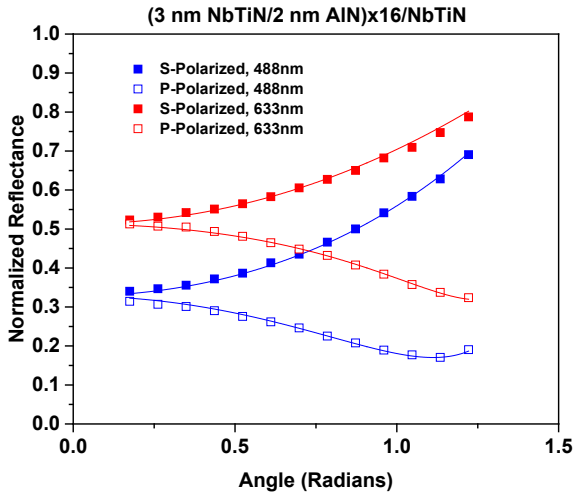

Fig. 9. Polarization reflectometry data for (3 nm NbTiN)/(2 nm AlN) x 16 multilayer. Solid lines are fits to Eqs. (1 – 4) with parameters:
$\varepsilon_{1\ (\text{in plane})} = -3.8 + 7.2i$
$\varepsilon_{2\ (\text{out of plane})} = 5.0$
for $\lambda = 633$ nm and
$\varepsilon_{1\ (\text{in plane})} = -0.51 + 4.7i$
$\varepsilon_{2\ (\text{out of plane})} = 1.3 + 7i$
for $\lambda = 488$ nm and

Polarization reflectometry showed (Fig. 9) that that 8 and 16-layered multilayers exhibit hyperbolic metamaterial behavior in the visible range with positive out-of-plane dielectric function and negative in-plane dielectric function.

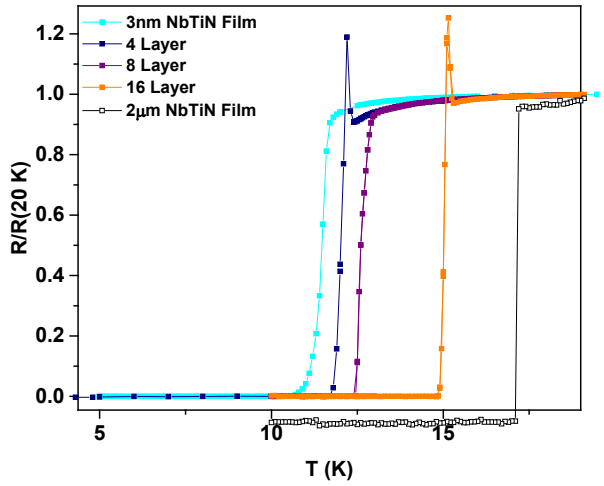

Fig. 10. Temperature dependence of normalized resistance of (3 nm NbTiN)/(2 nm AlN) multilayers and 2 micron thick single layer NbTiN film.

The superconducting critical temperature T_c increased with the number of layers in the multilayers, as it is expected as the metamaterial is formed from many layers (Fig. 10). Although the T_c of the multilayers is higher than the T_c of a single layer, it is lower than the T_c of a bulk material.

To determine whether T_c depends on a volume fraction of superconductor in our metamaterial, the thickness of dielectric were varied (Fig. 11). It was observed that for larger thicknesses of the dielectric layer (smaller volume fraction of superconductor) the T_c increases. The origin of this increase must be the **metamaterial effect**, since the other parameters (thickness of the NbTiN layer and number of layers) are kept the same.

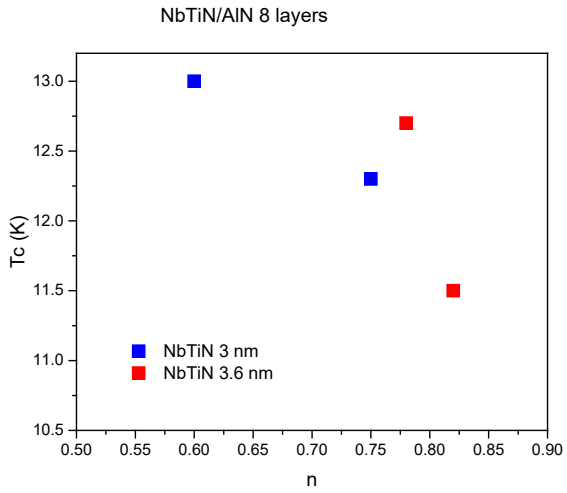

Fig. 11. The T_c of NbTiN/AlN 8-layers heterostructures on the volume fraction of superconductor.

Superconducting properties of tin-based ENZ and hyperbolic metamaterials

To better understand the potentials and limitation of the proposed metamaterial approach to enhancement of T_c, we performed a study of tin-based metamaterial superconductors in the epsilon-near-zero (ENZ) and hyperbolic metamaterial configurations. Our experimental results confirm recent theoretical predictions and earlier experimental observations that the superconducting critical temperature may be improved in various metamaterial superconductor geometries. Our detailed study of tin-based metamaterial superconductors in the epsilon-near-zero (ENZ) and hyperbolic metamaterial configurations demonstrated that the metamaterial superconductor approach remains valid even as the superconductor coherence length is strongly reduced compared to the very large value of ξ = 1600 nm in aluminum. It was observed that T_c enhancement is significantly reduced when the metamaterial structural dimensions exceed 240 nm, the superconducting coherence length in pure tin (Fig. 12). Smaller relative increases of T_c in tin-based metamaterial superconductors compared to similar aluminium-based metamaterial geometries may be explained by considerably larger imaginary part ε_m" of the tin dielectric constant. This work is published in Physica C: Superconductivity and its applications 565 (2019) 1353511.

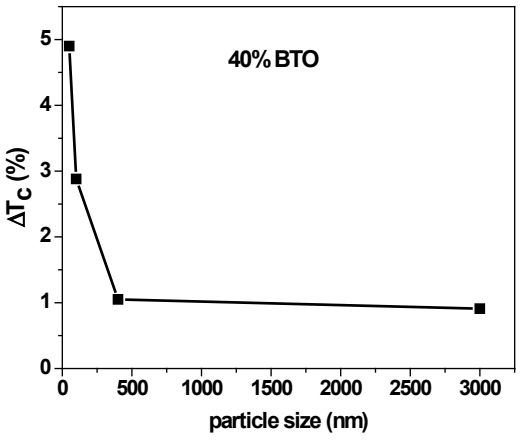

Fig. 12. The increase of T_c for the samples with 40% volume fraction of $BaTiO_3$ measured as a function of nanoparticle size. The superconducting critical temperature T_c increase appears to be strongly suppressed in metamaterial superconductors made with particle sizes considerably larger than the superconducting coherence length in pure tin ξ_{Sn} ~230 nm.

Theoretical

Coulomb blockade in the epsilon-near-zero regime

Theoretical efforts were directed toward refining the model of a hyperbolic metamaterial superconductor, which would go beyond their original estimates for the metamaterial parameters (unit layer thicknesses) in the MgB_2-based and Nb-based hyperbolic metamaterials. While the original estimates are based on relatively simple macroscopic electrodynamics approach, it was demonstrated that second order effects, such as the Coulomb blockade effect, need to be considered in the improved model. The new model demonstrates that similar to high T_c cuprates, hyperbolic metamaterial superconductors exhibit competition between superconductivity and charge ordering due to the Coulomb blockade effect.

On the other hand, enhancement of Coulomb blockade in the epsilon-near-zero regime (which also appears in hyperbolic metamaterials along certain directions) may lead to improved performance and/or relaxed nanofabrication requirements in optoelectronic Coulomb blockade devices, which is an important new result of our project.

The paper based on this work is published in Physica C: Superconductivity and its applications 556 (2019) 14–18.

Electron-electron pairing mediated by a hybrid plasmon-phonon excitation

To build our theoretical understanding of a mechanism of metamaterial enhancement of T_c, a theoretical model based on the Maxwell-Garnett effective medium approximation provides a microscopic explanation of this effect in terms of electron-electron pairing mediated by a hybrid plasmon-phonon excitation in the composite metal-dielectric metamaterial were developed. We report the first observation of a hybrid plasmon-phonon excitation in Al-Al_2O_3 ENZ core-shell metamaterials using inelastic neutron scattering (Fig. 13). These results provide strong support for this novel mechanism of superconductivity in ENZ metamaterials. The paper based on these findings is published in PHYSICAL REVIEW B **100**, 024515 (2019).

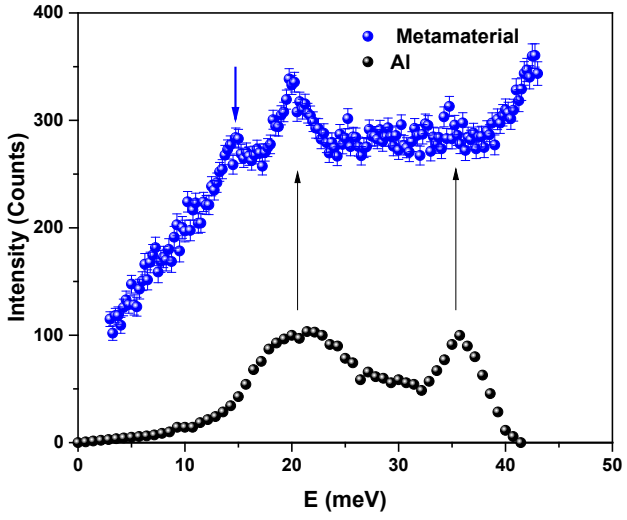

Fig. 13. Inelastic neutron scattering data for energies below 43 meV taken on BT-7, averaged over measurements taken at wave vectors Q = 4 Å^{-1}, 4.25 Å^{-1}, and 4.5 Å^{-1}. The data were taken at T = 5 K. The averaged dependence is proportional to the generalized phonon density of states (PDOS). The DOS for bulk aluminum at T = 10 K (black circles) is shown for comparison. Two peaks which are indicated with black arrows in the aluminium PDOS correspond to the van-Hove peaks for the transverse and longitudinal acoustical phonons, respectively, at the Brillouin zone boundaries. They are also present in the core-shell Al-Al_2O_3 metamaterial. The additional peak at around 15 meV (indicated with blue arrow) is not present in pure aluminum. It corresponds to the hybrid plasmon-phonons of the metamaterial.

Summary

We have used several methods to grow MgB_2/dielectric multilayers. However, the surface roughness of the MgB_2 layer prevents forming good quality multilayers with ultrathin layers of MgB_2: the layers are not well defined and conductance of MgB_2 is significantly lower. Nevertheless, the T_c in the attempted multilayers are higher than in a single layer of MgB_2 of corresponding thickness, which can be explained by metamaterial enhancement of T_c.

The multilayers of NbTiN/AlN of good quality were fabricated. These multilayers showed hyperbolic optical properties in the visible range. A steady increase of the T_c was observed with increase of the number of layers in the multilayers showing the onset of the metamaterial enhancement of T_c. The change of the T_c was observed when the thicknesses of the dielectric layer was increased while other parameters were kept constant. This also demonstrates that metamaterial parameters, like the volume faction of metal in the metamaterial, can enhance the T_c. However, the observed enhancement of the T_c did not exceed the T_c of the bulk NbTiN.

The reason for the limited enhancement of the T_c can be explained by the fact that the size of the metamaterial unit cell (the thickness of one bi-layer) is about the same as coherence length. Our work with tin-based metamaterial as a model system showed that T_c enhancement is significantly reduced when the metamaterial structural dimensions exceed the coherence length. Fabrication of superconductor/dielectric multilayers with smaller thickness of a bi-layer unit and at the same time having a good superconducting properties within the layer are needed.

We also report the first observation of a hybrid plasmon-phonon excitation in Al-Al_2O_3 ENZ core-shell metamaterials using inelastic neutron scattering. These results provide strong support for this novel mechanism of superconductivity in ENZ metamaterials.