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14. ABSTRACT
The DURIP program "Broadband Impedance Microscopy for Research on Complex Quantum Materials" supported by ARO has been successfully completed by the PI's group. By customizing the equipment purchased through this award, we have demonstrated the broadband impedance imaging with a spatial resolution better than 100 nm and a capacitance sensitivity better than 10 aF over 7 orders of magnitude in frequency (from 1 kHz to 10 GHz), as specified in the initially proposed program. In particular, exciting results were obtained to locally resolve the

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## Report Title

Final Report: Broadband Impedance Microscopy for Research on Complex Quantum Materials

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The DURIP program "Broadband Impedance Microscopy for Research on Complex Quantum Materials" supported by ARO has been successfully completed by the PI's group. By customizing the equipment purchased through this award, we have demonstrated the broadband impedance imaging with a spatial resolution better than 100 nm and a capacitance sensitivity better than 10 aF over 7 orders of magnitude in frequency (from 1 kHz to 10 GHz), as specified in the initially proposed program. In particular, exciting results were obtained to locally resolve the collective vibration of ferroelectric domain walls, which manifests itself as a frequency-dependent dielectric loss in the MHz-GHz regime. With the combined capability of dielectric spectroscopy and impedance microscopy, we expect to gain tremendous knowledge on complex quantum materials for their applications in the next-generation acoustic, electronic, and optic devices, all of which crucial for Defense purposes.

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## Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

### (a) Papers published in peer-reviewed journals (N/A for none)

Received Paper

**TOTAL:**

**Number of Papers published in peer-reviewed journals:**

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### (b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

**TOTAL:**

**Number of Papers published in non peer-reviewed journals:**

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### (c) Presentations

(1) "Near-field Microwave Impedance Microscopy on Novel Materials", 13th International Conference on Near-Field Optics, Denver CO, Sept 2014.
(2) "Nanoscale Impedance Imaging of Novel Quantum Materials", 2015 APS March Meeting, San Antonio TX, Mar 2015.
(3) "Broadband AC Conductivity in Ferroelectric Domain Walls", Nano, Polar, and Inorganic/Organic Materials Symposium, Philadelphia PA, Jun 2015.

**Number of Presentations:** 3.00

## Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

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Received Paper

**TOTAL:**

**Number of Manuscripts:**

## Books

Received Book

**TOTAL:**

Received Book Chapter

**TOTAL:**

## Patents Submitted

## Patents Awarded

## Awards

Keji Lai, International Union of Pure and Applied Physics (IUPAP) C10 Young Scientist Prize in the Structure and Dynamics of Condensed Matter, 2015

## Graduate Students

| NAME | PERCENT_SUPPORTED |
| --- | --- |
| **FTE Equivalent:** | |
| **Total Number:** | |

## Names of Post Doctorates

| NAME | PERCENT_SUPPORTED |
| --- | --- |
| **FTE Equivalent:** | |
| **Total Number:** | |

## Names of Faculty Supported

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| --- | --- |

**FTE Equivalent:**
**Total Number:**

## Names of Under Graduate students supported

| NAME | PERCENT_SUPPORTED |
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The number of undergraduates funded by this agreement who graduated during this period: ...... 0.00

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NAME

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## Names of personnel receiving PHDs

NAME

**Total Number:**

## Names of other research staff

| NAME | PERCENT_SUPPORTED |
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## Sub Contractors (DD882)

## Inventions (DD882)

## Scientific Progress

See Attachment

## Technology Transfer

# Final Report: Broadband Impedance Microscopy for Research on Complex Quantum Materials

Defense University Research Instrumentation Program (DURIP)

Army Research Office Grant #W911NF1410483

PI: Keji Lai, University of Texas at Austin, Department of Physics, Austin, TX 78712

List of illustrations:

## Statement of the Problem

The objective of this DURIP award is to construct a broadband impedance microscope (BIM) for frequency-dependent nanoscale imaging on complex quantum materials. Thanks to the ARO-DURIP support, we have acquired a capacitance/loss bridge (up to 20 kHz), an impedance analyzer (up to 50 MHz), a vector network analyzer (up to 43.5 GHz), and other peripheral electronics. Together with the existing scanning platforms and specialized cantilever probes [1] in the PI's lab, the BIM can now simultaneously obtain microscopic (10 – 100 nm) and quasi-spectroscopic (over 7 decades in frequency) information with unprecedented precisions. This novel experimental system will yield tremendous knowledge as we work toward building future acoustic, electronic, and optic devices for Defense applications.

Because of the cooperative interactions and competing orders, electronic inhomogeneity in the mesoscopic (nanometer to micrometer) length scale is widely observed in oxides and other complex systems [2-4]. Compared with single atoms or molecules, the collective motion of mesoscopic domains or domain walls is relatively slow, with characteristic frequencies ranging from quasi-static to the microwave regime. As seen in Fig. 1, macroscopic dielectric spectroscopy experiments have been done on various bulk materials, showing the typical dielectric relaxation and resonant behaviors [5-7]. Little is known, however, on the microscopic details of these processes.

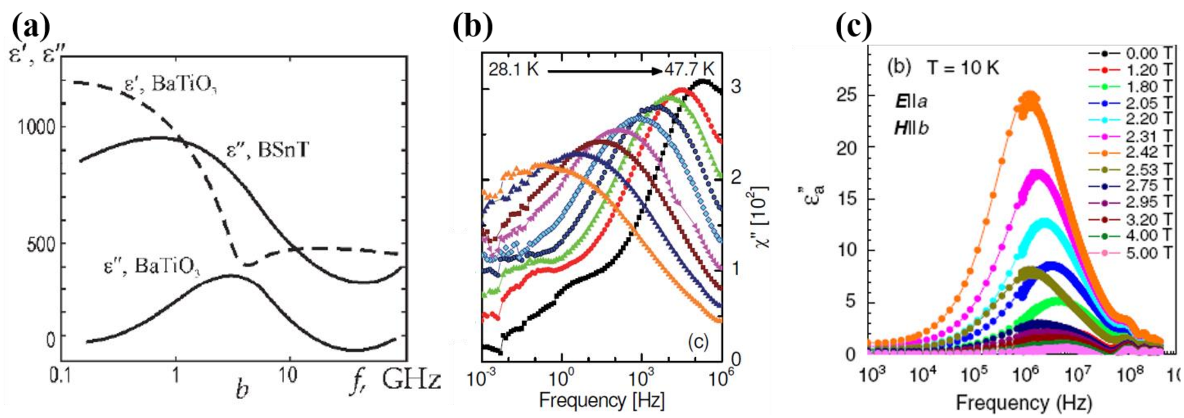


Fig. 1. Frequency-dependent dielectric function in various materials. (a) Real (ε′) and imaginary (ε″) components of ferroelectric $BaTiO_3$ from 100 MHz to 100 GHz. A strong relaxation peak is observed in ε″ around 3 GHz [5]. (b) Imaginary part of the relative dielectric susceptibility (χ″ = ε″) of doped incipient ferroelectric $Sr_{0.98}Mn_{0.02}TiO_3$ between 28 K and 48 K [6]. (c) ε″ of $DyMnO_3$ from zero field to B = 5 T, showing a strong relaxation peak at ~1 MHz [7].

Before our work, it was technically difficult to obtain nanoscale electrical properties for a broad range of frequencies. As one goes from a typical bulk sample with 1 mm in size to a modest spatial resolution of 0.1 μm, the volume being probed drops by a factor of $10^{12}$, and so does the measured signal. In addition, high-frequency measurements are notoriously susceptible to artifacts due to stray field contribution. The shielded cantilever probes [1] developed by the PI proves to be crucial for any meaningful effort. With this ARO-DURIP grant, the PI's group was able to purchase several state-of-the-art electronics, each covering 3 ~ 4 decades of frequency for best sensitivity and stability, to take on this challenge task.

**Summary of Key Results**

The configuration of the BIM is detailed as follows. A summary of the frequency-dependent sensitivity of the microscope is shown in Fig. 2. An instrumentation paper is currently in preparation to describe the BIM.

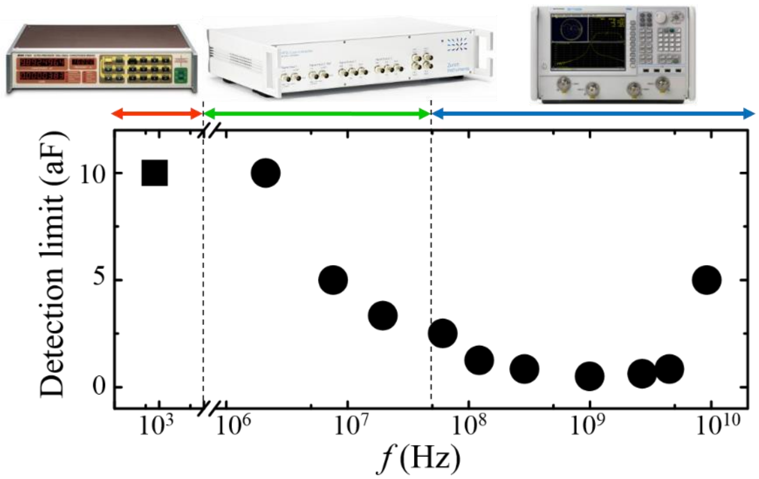


Fig. 2. Sensitivity limit in terms of changes in the tip capacitance of the broadband impedance microscope (BIM). The three instruments (from left to right), capacitance/loss bridge AH2700A, high-f lock-in amplifier HF2LI, and PNA microwave network analyzer N5244A, can cover the broad frequency spectrum between 1 kHz and 10 GHz for the impedance detection.

a. For the low-frequency or quasi-static range, we purchased the capacitance/loss bridge AH2700A from Andeen-Hagerling Inc., which offers continuous frequency operation from 50 Hz to 20 kHz, with the best sensitivity of ~ 1 aF achieved at 1 kHz. This is, however, only achieved with a very long averaging time that is not compatible with scanning probe experiments. We have confirmed that, in practice, the sensitivity is close to 10 aF with an integration time of ~ 20 ms.

b. For frequencies up to 50 MHz, we acquired the HF2LI lock-in amplifier with the multiple-frequency option and reconfigured it with other peripheral components into a sensitive impedance detector. With careful impedance match to the cantilever probe, we have demonstrated a capacitance detection limit below 10 aF with the normal scanning rate (about 20 min per frame). Note that the commercial Precision Impedance Analyzers, e.g., Keysight 4294A, only guarantee a detection limit on the order of 100 aF, which is clearly inferior to our design.

c. For frequencies up to 20 GHz, we procured the N5244A PNA Microwave Network Analyzer from Keysight Technologies Inc. Using similar impedance-match schemes and peripheral circuits, we again showed that a capacitance change as small as 1 – 10 aF can be readily detected with 1 kHz bandwidth. Note that while the electronics during our preliminary testing only went up to ~ 10 GHz, the analyzer can potentially reach 43.5 GHz for reflectivity measurements.

The implementation of the BIM has found immediate applications in complex systems. Many ferroelectric materials, which are widely used in communication systems and electrically controlled devices such as varactors and phase shifters, exhibit a strong dielectric dispersion in the microwave regime [8]. While the response is commonly attributed to the translational vibrations of domain walls [9], there have not been any microscopic studies down to the single domain wall level.

Fig. 3 shows our preliminary BIM data on the z-cut surface of the improper ferroelectric hexagonal manganite $YMnO_3$. In order to minimize the electrostatic energy, ferroelectric and structural antiphase boundaries are mutually interlocked, with six domain walls merging into a single vortex core and displaying a beautiful cloverleaf pattern [10]. Previous conductive atomic-force microscopy (C-AFM) work has shown that the dc conductivity of the domain walls in $YMnO_3$ is very low (<< 1 S/m) [11]. Surprisingly, clear domain wall contrast was observed for a broad frequency range from 1 MHz to 10 GHz. Using finite-element analysis tools [12], we can further quantify the ac conductivity of the domain walls to be $10^2$ – $10^3$ S/m, which is 5 ~ 6 orders of magnitude higher than that the dc value. While the underlying physics is not fully understood, the BIM experiment clearly reveals a highly nontrivial dynamic response of the coupled walls to the oscillating electric fields. A manuscript is currently in preparation to report this exciting discovery in a high-impact journal.

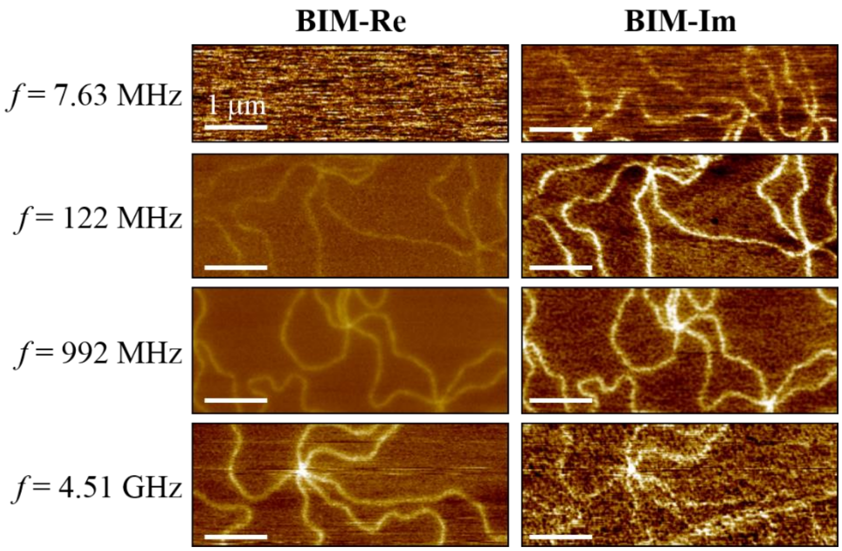


Fig. 3. Preliminary BIM data on $YMnO_3$. The real (BIM-Re) and imaginary (BIM-Im) output channels of the BIM images are displayed here with the same false-color scale. The cloverleaf patterns of the domain wall structures are observed, indicative of the strong ac conductivity of the walls. All scale bars are 1 μm.

To summarize, thanks to the DURIP support, the PI's group has acquired the necessary electronics to perform nanoscale impedance imaging over a broad electromagnetic spectrum. The sensitivity of the instrument is characterized to be better than 10 aF over 7 orders of magnitude ($10^3$ to $10^{10}$ Hz) in frequency. Our preliminary BIM data on ferroelectric materials revealed highly nontrivial electromagnetic responses of the domain walls. We expect that this unique tool will make substantial contributions to the vibrant field of condensed matter physics. The research to be conducted in the near future based on the BIM is well aligned with the ARO mission to understand emergent phenomena in strongly correlated systems and information in non-equilibrium nanosystems.

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