

The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA, 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.
PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.

1. REPORT DATE (DD-MM-YYYY) 07-04-2019	2. REPORT TYPE Final Report	3. DATES COVERED (From - To) 1-Aug-2014 - 31-Dec-2018
---	--------------------------------	--

4. TITLE AND SUBTITLE Final Report: Quantum Critical Behavior in Oxide Structures	5a. CONTRACT NUMBER W911NF-14-1-0379
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER 611102

6. AUTHORS	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES University of California - Santa Barbara 3227 Cheadle Hall 3rd floor, MC 2050 Santa Barbara, CA 93106 -2050	8. PERFORMING ORGANIZATION REPORT NUMBER
--	--

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211	10. SPONSOR/MONITOR'S ACRONYM(S) ARO
	11. SPONSOR/MONITOR'S REPORT NUMBER(S) 65675-PH.21

12. DISTRIBUTION AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.
--

13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:	17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON Susanne Stemmer
a. REPORT UU	b. ABSTRACT UU	c. THIS PAGE UU	19b. TELEPHONE NUMBER 805-893-6128

RPPR Final Report
as of 14-May-2019

Agency Code:

Proposal Number: 65675PH

Agreement Number: W911NF-14-1-0379

INVESTIGATOR(S):

Name: Susanne Stemmer
Email: stemmer@mrl.ucsb.edu
Phone Number: 8058936128
Principal: Y

Organization: **University of California - Santa Barbara**

Address: 3227 Cheadle Hall, Santa Barbara, CA 931062050

Country: USA

DUNS Number: 094878394

EIN: 956006145W

Report Date: 31-Mar-2019

Date Received: 07-Apr-2019

Final Report for Period Beginning 01-Aug-2014 and Ending 31-Dec-2018

Title: Quantum Critical Behavior in Oxide Structures

Begin Performance Period: 01-Aug-2014

End Performance Period: 31-Dec-2018

Report Term: 0-Other

Submitted By: Susanne Stemmer

Email: stemmer@mrl.ucsb.edu

Phone: (805) 893-6128

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

STEM Degrees: 4

STEM Participants: 7

Major Goals: The objective of the project "Quantum Critical Behavior in Oxide Structures" was to advance the understanding of quantum criticality involving delocalized electrons in strongly correlated systems. We took advantage of the control afforded by oxide heterostructures and rationally design and manipulate quantum criticality. Different types of interactions were introduced and have led to an understanding of their relevance in driving quantum critical phenomena and in the promotion of emergent ordered states, thereby enabling the development of a predictive theory.

Accomplishments: Please see attachment and also the publication list.

Training Opportunities: Several graduate students worked on this project. They were trained in advanced many body theory, neutron, synchrotron x-ray, and muon techniques, thin film growth by molecular beam epitaxy and electrical measurements. Results from the project contributed to their Ph.D. theses.

Results Dissemination: Results were disseminated via journal publications (submitted on this site) and conference/workshop talks given by PIs and their students.

Honors and Awards: Ryan Need was the recipient of a postdoctoral fellowship from the National Research Council.

Balents was elected a Fellow of the American Academy of Arts and Sciences. Stephen Wilson received a Hellman Faculty Fellowship

Stemmer received a National Security Science and Engineering Faculty Fellowship (Vannevar Bush Faculty Fellowship)

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: Other Professional

Participant: Maxim METLITSKI

Person Months Worked: 3.00

Funding Support:

RPPR Final Report
as of 14-May-2019

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Other Professional

Participant: Thomas Hogan
Person Months Worked: 4.00

Funding Support:

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Co PD/PI

Participant: Leon Balents
Person Months Worked: 2.00

Funding Support:

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: PD/PI

Participant: Susanne Stemmer
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: Stephen Wilson
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: Kaveh Ahadi
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: Kara HEJAZI

RPPR Final Report
as of 14-May-2019

Person Months Worked: 6.00

Funding Support:

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: Jason IACONIS

Person Months Worked: 4.00

Funding Support:

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: CHUNXIAO LIU

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: Evgeny Mikheev

Person Months Worked: 2.00

Funding Support:

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

Participant Type: Graduate Student (research assistant)

Participant: Santosh Raghavan

Person Months Worked: 3.00

Funding Support:

Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

ARTICLES:

RPPR Final Report as of 14-May-2019

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: ACS Nano

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acsnano.6b08427

Volume: 11

Issue: 4

First Page #: 3760

Date Submitted: 12/4/17 12:00AM

Date Published: 3/1/17 8:00AM

Publication Location:

Article Title: Potential Fluctuations at Low Temperatures in Mesoscopic-Scale SmTiO

Authors: Will J. Hardy, Brandon Isaac, Patrick Marshall, Evgeny Mikheev, Panpan Zhou, Susanne Stemmer, Dou

Keywords: Non-Fermi liquids, oxide quantum wells

Abstract: Heterointerfaces of SrTiO₃ with other transitionmetal oxides make up an intriguing family of systems with a bounty of coexisting and competing physical orders. Some examples, such as LaAlO₃/SrTiO₃, support a high carrier density electron gas at the interface whose electronic properties are determined by a combination of lattice distortions, spin-orbit coupling, defects, and various regimes of magnetic and charge ordering. Here, we study electronic transport in mesoscale devices made with heterostructures of SrTiO₃ sandwiched between layers of SmTiO₃, in which the transport properties can be tuned from a regime of Fermi-liquid like resistivity ($\rho \propto T^2$) to a non-Fermi liquid ($\rho \propto T^{5/3}$) by controlling the SrTiO₃ thickness.

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

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Reports on Progress in Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1088/1361-6633/aabdfa

Volume: 81

Issue: 6

First Page #: 062502

Date Submitted: 6/4/18 12:00AM

Date Published: 6/1/18 7:00AM

Publication Location:

Article Title: Non-Fermi liquids in oxide heterostructures

Authors: Susanne Stemmer, S James Allen

Keywords: Non-Fermi liquids, oxide heterostructures, quantum criticality

Abstract: Understanding the anomalous transport properties of strongly correlated materials is one of the most formidable challenges in condensed matter physics. For example, one encounters metal-insulator transitions, deviations from Landau Fermi liquid behavior, longitudinal and Hall scattering rate separation, a pseudogap phase, and bad metal behavior. These properties have been studied extensively in bulk materials, such as the unconventional superconductors and heavy fermion systems. Oxide heterostructures have recently emerged as new platforms to probe, control, and understand strong correlation phenomena. This article focuses on unconventional transport phenomena in oxide thin film systems. We use specific systems as examples, namely charge carriers in SrTiO₃ layers and interfaces with SrTiO₃, and strained rare earth nickelate thin films.

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

Acknowledged Federal Support: Y

RPPR Final Report as of 14-May-2019

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review Materials

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevMaterials.2.093801

Volume: 2

Issue: 9

First Page #: 093801

Date Submitted: 9/6/18 12:00AM

Date Published: 9/1/18 7:00AM

Publication Location:

Article Title: Resolving interfacial charge transfer in titanate superlattices using resonant x-ray reflectometry

Authors: R. F. Need, P. B. Marshall, E. Weschke, A. J. Grutter, D. A. Gilbert, E. Arenholz, P. Shafer, S. Stemmer

Keywords: Oxide Interfaces, X-ray reflectivity,

Abstract: Charge transfer in oxide heterostructures can be tuned to promote emergent interfacial states, and accordingly, has been the subject of intense study in recent years. However, accessing the physics at these interfaces, which are often buried deep below the sample surface, remains difficult. Addressing this challenge requires techniques capable of measuring the local electronic structure with high-resolution depth dependence. Here, we used linearly polarized resonant x-ray reflectometry (RXR) as a means to visualize charge transfer in oxide superlattices with single unit cell precision. From our RXR measurements, we extract valence depth profiles of SmTiO₃ (SmTO)/SrTiO₃ (STO) heterostructures with STO quantum wells varying in thickness from five SrO planes down to a single SrO plane.

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

Acknowledged Federal Support: Y

Accomplishments

The objective of the project “*Quantum Critical Behavior in Oxide Structures*” was to advance the understanding of quantum criticality involving delocalized electrons in strongly correlated systems. The project took advantage of the control afforded by oxide heterostructures and rationally design and manipulate quantum criticality. Different types of interactions were introduced and led to an understanding of their relevance in driving quantum critical phenomena and in the promotion of emergent ordered states, thereby enabling the development of predictive theories. Below we summarize the progress in this project over the first year of the project.

A major focus of the experimental and theoretical work in the project was on two-dimensional electron liquids in quantum well structures of type $RTiO_3/SrTiO_3/RTiO_3$ ($R = Gd$ or Sm), which contain a high mobile charge density in the $SrTiO_3$ quantum well. The $RTiO_3$ are prototype Mott insulators. These structures were used as a model system to explore questions such as the origins of quantum criticality, and associated phenomena (pseudogaps, non-Fermi liquid behavior,...) and their tunability in oxide heterostructures.

Using molecular beam epitaxy (MBE), the **Stemmer** group grew two-dimensional electron liquids (2DELs) in quantum well structures of type $RTiO_3/SrTiO_3/RTiO_3$ ($R = Gd$ or Sm). A major finding was that the transport data could be modeled using separate transport and Hall scattering rates, also known as “two-lifetime” behavior. This framework gave a remarkably simple and general description of the temperature dependence of the Hall coefficient. We showed that the 0-K Hall coefficient diverged at a critical quantum well thickness, coinciding with a quantum phase transition. We found that the Hall angle follows a T^2 temperature dependence, even when the longitudinal resistance does not. The clear separation the residuals supports the notion of distinct scattering rates in these systems, with different underlying physics that influences them. The data pointed to fluctuations associated with an underlying quantum critical point (QCP), which are reflected in the 0-K Hall coefficient, as the origin of the two lifetime behavior.

We also investigated the influence of disorder on the “two-lifetime” behavior. Disorder can enhance the effects of electron correlations and give rise to non-Fermi liquid behavior. Introducing controlled amounts of disorder can assist in elucidating the transport physics in correlated materials. Disorder was introduced via planar growth defects normal to the quantum wells, which lead to strongly anisotropic transport properties. Using angle-dependent sheet resistance and Hall measurements, the contributions of the defects and those of intrinsic scattering mechanisms to the transport properties and lifetime separation were determined. The residuals in both the longitudinal resistance and Hall angle were found to depend on the relative orientations of the transport direction to the planar defects. The Hall angle exhibited a robust T^2 temperature dependence along all directions, whereas no simple power law could describe the temperature dependence of the longitudinal resistances. Remarkably, the degree of the carrier lifetime separation, as manifested in the distinctly different temperature dependences and diverging residuals near a critical quantum well thickness, was completely insensitive to disorder. We speculated that the robust T^2 behavior of the Hall angle reflects a universal, underlying scattering rate.

One of the key unknowns in driving and ultimately controlling quantum criticality is the nature of the order parameter. To this end, the **Wilson** group focused on directly resolving the nature of the (magnetic) order parameter underlying the QCP of this system. A major activity

centered on experimentally interrogating the role of the *spin degree of freedom* in quantum critical magnetotransport behavior. All samples were grown by molecular beam epitaxy (MBE) in the **Stemmer** group.

As a starting point, the **Wilson** group focused on the magnetization density of quantum well heterostructures. Neutron reflectometry was used to directly resolve how magnetization density induced via the neighboring GdTiO_3 is distributed across the quantum wells (where the electrons flow in charge transport). The data showed that the entirety of the quantum well is magnetized at low temperatures and charge transport necessarily pushes through a volume of magnetic SrO .

As a next step, the **Wilson** group explored the correlation between the onset of long-range magnetic order or incipient magnetic fluctuations and the appearance of transport phenomena coupled to quantum criticality. Features of particular interest included the emergence of a pseudogap state, discovered by the **Stemmer** group, and divergent carrier mass near a critical quantum well thickness in $\text{SmTiO}_3/\text{SrTiO}_3$ heterostructures, as well as the underlying coupling between magnetism and octahedral distortions in $\text{GdTiO}_3/\text{SrTiO}_3$ heterostructures.

The **Wilson** group performed low energy muon implantation measurements on an $\text{SmTiO}_3/\text{SrTiO}_3$ heterostructure, as well as on a pristine SmTiO_3 film. In these measurements, they were able to resolve the onset of antiferromagnetism in both the Ti and Sm sublattices within the isolated SmTiO_3 film. These are the first measurements able to detect magnetism within an ultrathin (~ 20 nm thick) SmTiO_3 film—a lingering question given the reduced structural symmetry of the thin films relative to bulk crystals. More importantly, the measurements of a $\text{SmTiO}_3/\text{SrTiO}_3/\text{SmTiO}_3$ trilayer heterostructure revealed that an additional channel of spin freezing appears below $T^* \sim 20$ K after both the Sm and Ti sublattices have ordered in the SmTiO_3 layers. This new channel of freezing represents the onset of incipient antiferromagnetic order within the SrTiO_3 quantum wells and it coincides with the prior observation of a pseudogap opening in this system. The measurements connect the formation of the pseudogap state in high-density $\text{SmTiO}_3/\text{SrTiO}_3$ quantum wells with emergence of antiferromagnetic correlations within the wells. More broadly, the work ties the pseudogap formation with previous observations of pseudogaps in carrier-tuned bulk Mott insulating systems such as high- T_c cuprates and manganites.

A second major activity in the **Wilson** group concerned the exploration of the coupling between the octahedral tilts and ferrimagnetism within GdTiO_3 layers. As GdTiO_3 is interfaced with SrTiO_3 , the TiO_6 octahedral tilts at the interface are perturbed, and for thin GdTiO_3 layers constrained within thicker SrTiO_3 layers, these tilts are suppressed toward the undistorted lattice of SrTiO_3 . As the tilts are relaxed within the GdTiO_3 layers, the apparent magnetic moment (as seen via bulk magnetometry) associated with ferromagnetism is suppressed. At the thin layer limit, previous magnetometry work was unable to resolve any ferrimagnetism in GdTiO_3 , suggesting that the magnetic moment of Ti is substantially renormalized when the GdTiO_3 octahedral tilts are relaxed. Our work exploring this effect utilized polarized neutron reflectometry to uncover two key findings. The first is that this apparent suppression is due to the formation of magnetic deadlayers within the GdTiO_3 films once interfaced with SrTiO_3 . The second is that, once these deadlayers are accounted for, the ferrimagnetic moment of GdTiO_3 was found to be unaffected by relaxed TiO_6 tilts in thin GdTiO_3 layers. Curiously, this demonstrates that the magnetism is unaffected by the large modifications of the octahedral tilts, and presumably the t_{2g} bandwidth, and suggests that our theoretical understanding of magnetism in this system should be revisited.

The theoretical efforts in the project (**Balents** group) focused on understanding the *origin* of quantum criticality in titanate heterostructures. The experiments of the **Stemmer** and **Wilson** groups pointed to essential differences of the heterostructures involving GdTiO_3 and SmTiO_3 (in particular with respect to the magnetism and non-Fermi liquid behavior), which suggests that the nature of the Mott insulator is an important ingredient in the types of orders and quantum phase transitions which occur. Pursuant to this idea, the **Balents** group constructed a minimal model of the Mott insulator to band insulator interface. Crucially, in addition to the 2DEL induced by the polar discontinuity inside the band insulator (SrTiO_3), the model takes into account the very high density (1 electron per Ti) of *localized* electrons inside the Mott material. While these do not directly contribute to transport, they carry a large amount of entropy and can strongly influence magnetism and quantum criticality.

The model can be *solved* in several regimes in a very controlled fashion, using both analytical approaches and the numerically *exact* method of Density Matrix Renormalization Group (DMRG). The main finding is that the proximity to the Mott layer strongly induces magnetism, which can be either ferromagnetic or antiferromagnetic, depending upon details of carrier density, band offset, etc. The mechanism for these orderings is *kinetic* in nature: electrons which are coupled to the Mott layer move more coherently in a specific magnetic background. Oxide heterostructures might in fact be the first experimental realization of this idea, which was first proposed (in a simpler and rather different context) many decades ago. The model predicts a large range of ferromagnetism and competing antiferromagnetic states at lower electron density.

Theoretical research in the **Balents** group also aimed developing a theoretical description of non-Fermi liquid behavior, especially transport, in strongly correlated metals, such as those arising at quantum critical points. Specifically, they sought a description outside the usual weak-scattering approaches and distinct from Hertz-Millis-Moriya theory, which retains the quasiparticle basis and does not seem to apply to many experimental systems. Furthermore, they sought to understand microscopically how all these phenomena arise from details of electronic orbitals, geometry, and Kanamuri interactions. The main theoretical output was to develop a new approach to non-Fermi liquids starting from what is now called the Sachdev-Ye-Kitaev (SYK) model, which became the subject of intense study as a toy model in which gravity and black hole physics emerges from totally different microphysics: quantum mechanics of many fermions. It has a very simple Hamiltonian involving very complicated interactions that create transitions from two “orbitals” (kl) to another two (ij) in pairs, amongst N total orbitals. The SYK model is one of a very few solvable theoretical models for non-Fermi liquids, which makes it quite valuable. The drawback of the SYK model is that it is “zero dimensional” — every electron in it interacts with every other one, and so there is no built-in locality to the problem.

What the **Balents** group did was to string together these dots as super-atoms, connecting them by one-electron hopping. In this model, there is a competition between the SYK interaction U and the hopping t . For $t \ll U$, we have a strongly correlated system. It remains fully soluble in the large N limit and one can obtain a rich structure. Briefly, at low temperatures and low energy, the system is a Fermi liquid. This allows one to extract the parameters of the Fermi liquid theory: effective mass, Fermi liquid interactions, quasiparticle residue, etc. However, at higher temperatures it is an incoherent metal, and displays properties that are a mixture of those of the pure SYK model and a regular metal. Together, it is striking how many of the properties are similar to those of experimental correlated metals:

$$\text{Small coherence scale } E_c = t^2/U$$

Large effective mass (Sommerfeld coefficient) $\gamma \sim m^*/m \sim U/t$

Small quasiparticle weight $Z \sim t/U$

T^2 low-temperature resistivity with Kadowaki-Woods ratio $A/\gamma^2 = \text{constant}$

linear in temperature resistivity at high temperature

linear in temperature thermal “resistivity” T/κ at high temperature

Fermi liquid Lorenz number $L = \kappa/(T \sigma) = \pi^2/3$ at low T

non-Fermi liquid Lorenz number $L = \pi^2/8$ at high T