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6. AUTHORS
JIGANG WANG

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES
Iowa State University of Science and Techn
1138 Pearson Hall
505 Morrill Road
Ames, IA 50011 -2103

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14. ABSTRACT
This workshop will critically examine the state-of-the-art, envision the potential for far-reaching discoveries, and stimulate novel concepts and proposals in the emerging frontier of ultrafast quantum materials science. In view of the significant recent breakthrough of femtosecond non-equilibrium approach to strongly-correlated electrons and low-dimensional quantum fluids, the workshop represents an opportunity for a paradigm shift in quantum phase discovery and understanding with a strong long-term impact on Army fundamental research needs and some grand

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Report Title

Final Report: ARO Workshop on Non-Equilibrium Quantum Matter and Phase Transitions Created by Strongly-Correlated Ultrafast Excitations

ABSTRACT

This workshop will critically examine the state-of-the-art, envision the potential for far-reaching discoveries, and stimulate novel concepts and proposals in the emerging frontier of ultrafast quantum materials science. In view of the significant recent breakthrough of femtosecond non-equilibrium approach to strongly-correlated electrons and low-dimensional quantum fluids, the workshop represents an opportunity for a paradigm shift in quantum phase discovery and understanding with a strong long-term impact on Army fundamental research needs and some grand challenges. Particularly, the questions and discussions for the proposed workshop will be driven by a vision that by selectively manipulating quantum states with laser-driven non-adiabatic dynamics and coherent excitations, versatile ultrafast spectroscopic tools will reveal fundamental emergent-order phenomena via discovering and engineering hidden phases in highly non-thermal, non-equilibrium states. One major goal is to bring some of the leading experts, both experimental and theoretical, from complementary areas to discuss bold schemes and to advance this tough, intellectually stimulating question to push the field forwards.

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)

<u>Received</u> <u>Paper</u>

TOTAL:

Number of Papers published in peer-reviewed journals:

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

<u>Received</u> <u>Paper</u>

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Number of Presentations: 0.00

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

Received   Paper

TOTAL:

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

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

Received   Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received   Paper

TOTAL:

Number of Manuscripts:

Books

Received   Book

TOTAL:

Received Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED

FTE Equivalent:
Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

See attachment

Technology Transfer

Report on ARO workshop on Non-equilibrium Quantum Matter Created by Strongly-Correlated Ultrafast Excitations

Jigang Wang
Iowa State University and Ames Laboratory

Overview: On December 11-12, 2014, a small group of leading experts, both experimental and theoretical, from complementary ultrafast non-equilibrium physics areas participated in an ARO workshop entitled "Non-equilibrium Quantum Matter Created by Strongly-Correlated Ultrafast Excitations". They were charged to critically examine the state-of-the-art, outstanding questions, envision the potential for far-reaching discoveries, and stimulate novel concepts and proposals in the emerging frontier of ultrafast quantum materials science. The bulk of the workshop was spent in both invited presentations and discussion sessions. The scientific programs were exciting and the discussions were intense and productive. Dr. Marc Ulrich from the Army Research Office motivates the participants on identify the elements of the most compelling scientific concepts and technical capabilities for the non-equilibrium quantum materials research. The workshop speakers rose to the questions effectively, deliberated several bold schemes and identified stimulating questions which will set the agenda for their own research in the years ahead. These will influence the whole community through their work in the forefront of this new discipline. The new opportunity in quantum phase discovery and understanding will also have a strong long-term impact on Army fundamental research needs and some grand challenges. For example, the recurring theme targets "probing and controlling unique phenomena, especially in the studies of novel quantum phases of matter", "information processing/communications", "ultrafast electronics, optoelectronics", and "complex oxide heterostructures" which are all important thrusts specifically identified in the ARO research profile. A list of outstanding questions are recorded below which provides a framework for recommendations of compelling conceptual and technical capabilities in the end.

Scientific background: Traditionally, exotic quantum phases in advanced materials have been discovered and studied by thermodynamic tuning with chemical substitution, high pressure or strong magnetic field. The order parameters of these equilibrium phases are a manifestation of spontaneous coherence, e.g. between many-body states separated by the Mott–insulator energy gap or pairing electrons with the off-diagonal long range order, mediated by some low energy charge/spin fluctuations. In the ultrafast perturbation and/or coherent regime, one can expect to generate forbidden mixtures of order parameters and hidden metastable phases since the system does not have time to respond to the changes in the Hamiltonian and will depart from the instantaneous ground state. *This out-of-equilibrium spectroscopy approach* provides a "time-window of opportunity" for discovering exotic quantum phenomena and hidden quantum phases. In parallel, ultrafast measurement techniques, which span an extraordinary dynamical range using various complementary probes, both photons, from terahertz to x-ray spectral regions, and electron beams, allows to simultaneously monitor various components and fluctuations of complex order parameters.

Issues identified in non-equilibrium quantum matter: Some outstanding and cross-cutting questions are listed below which provides a framework for recommendations of compelling conceptual and technical capabilities next.

- Is it possible to identify and characterize true broken symmetry phenomena in quantum materials far from equilibrium?
- Can one identify some nearly-closed, quantum correlated condensed matter systems that are far from equilibrium?
- Is it possible to manipulate quantum coherence and entanglement at the macroscopic level and ultrafast time scales?
- How does the hidden state in the quantum materials arise from a normal, equilibrium state after ultrafast photoexcitations? What are the formation pathways?
- What are the dynamical signatures at finite temperatures associated with quantum critical points (they mark zero-temperature phase transitions driven by quantum fluctuations) in iron pnictide superconductors?
- How to manipulate spin, charge and orbital ordering in transition-metal oxides by mode-selective excitations?
- How to achieve ultrafast control of quantum magnetism and identity salient features of non-adiabatic spin dynamics in model systems e.g., those exhibit spin-nematicity?
- What is the structure, properties and function relationship in ultrafast laser-induced phase transitions in strongly correlated metal-oxide systems, e.g., V-, Mn- and Co-…?

Compelling conceptual and technical capabilities recommended: The ultrafast non-equilibrium approach becomes an excellent avenue for the investigation of the questions above and complex materials. The process must be driven by the both new concepts and creative way of using technical capabilities.

- Develop quantum excitation protocols that enable creating long-lived metastable quantum states and hidden orders, which provides a complementary way to conventional materials fabrication/discovery.
- Develop reliable and robust instrumentations with simultaneous space, energy and time visualization, i.e., nanometer in space, femtosecond in time and THz in energy, at a single quanta level (1-electron, 1-photon, 1-spin).
- Combine the ultrafast instrumentations with the control knobs similar to those used by the traditional condensed matter/materials physics community such as high magnetic field, high pressure and ultra-low temperature, and, yet, to provide the addition of fs time resolution.
- Correlate various advanced, complementary techniques to study the same model systems, e.g., THz pump/THz probe, ultrafast x-ray and electron spectroscopy techniques, ultrafast magneto-optical techniques
- Develop theoretical simulations of the actual experimental results and make further predictions to guide the experiments.

Further expand the impacts of the workshop: The discussions and ideas initiated by in the workshop help develop proposals, initiate collaborations and join papers.

- The W.M. Keck Foundation recently awarded Dr. J. Wang a three-year grant to support construction, commissioning and initial use of an ultrafast THz nanoscope.
- The discussions in the workshop have partially helped two papers, one submitted and the other in final preparation:
  1. A. Patz, et al., Critical speeding-up of non-equilibrium electronic relaxation near Nematic phase transition in unstrained $Ba(Fe_{1-x}Co_x)_2As_2$, in review, Phys. Rev. B (2016)
  2. L. Luo, et al., Non-thermal fs generation of hidden spin order in $LaCoO_3$, in preparation, Phys. Rev. Lett (2016)

  Proper acknowledgements to this grant have been included in these papers.