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RPPR Final Report
as of 11-Feb-2019

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Report Date: 10-Dec-2018 Date Received: 13-Jan-2019
Final Report for Period Beginning 11-Sep-2017 and Ending 10-Sep-2018

Title: Acquisition Of an Ultrafast Amplified Laser System for Research and Education in Material Science

Begin Performance Period: 11-Sep-2017 **End Performance Period:** 10-Sep-2018
Report Term: 0-Other

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STEM Degrees: 0 **STEM Participants:** 3

Major Goals: The major goal of this project is to acquire an ultrafast amplified laser system at Florida International University to enhance our capabilities in material science research and education.

The ultrafast laser system consists of the following items:

a. A femtosecond, kHz one-box amplifier system (Coherent Astrella),
b. A femtosecond Optical Parametric Amplifier (OPA) with the second harmonic and sum frequency option (Coherent OPerA with the SF package),
c. A femtosecond OPA with the nonlinear different frequency generation option (Coherent OPerA with the NDFG package),
d. An optical table with an isolation system.

The one-box, femtosecond amplifier system (Astrella) is an integrated amplifier system including a femtosecond oscillator (Vitara), a pulsed pump laser (Revolution), a regenerative amplifier and a stretcher/compressor. Astrella produces 7-mJ, 35-fs pulses at a repetition rate of 1 kHz and a center wavelength of 800 nm. The output of Astrella will be used to simultaneously drive two OPAs (OPerA) that extend the wavelength coverage. Both OPerAs have a Signal beam (1140~1600 nm) and an Idler beam (1600~2600 nm). The FH package in the first OPerA has a tuning range from 480~533 nm for the SFS beam, 533~580 nm for the SFI beam, 580 to 800 nm for the SHS beam, and 800 to 1150 nm for the SHI beam. The output pulse energy is >30 uJ. The NDFG package in the second OPerA can be tuned from 2.6 to 9 um. The output pulse energy is >2 uJ at 4 um and >0.5 uJ at 9 um. The pulse duration for the outputs of both OPerAs is shorter than 53 fs. The whole laser system will be installed on an isolated optical table.

The ultrafast laser system was expected to be the first and only ultrafast amplifier laser system at Florida International University (FIU). The system can provide FIU researchers with new capabilities in three aspects: ultrashort pulses (<35 fs), high pulse energy (up to 7 mJ), and broad wavelength coverage (480 nm ~ 9 um). These new capabilities will be integrated with the existing material research programs in four academic departments at FIU and enhance the research in material science and ultrafast spectroscopy. The system will enhance FIU's education in the STEM fields by providing unique research opportunities that are unprecedented to FIU students, including underrepresented minority groups.

Accomplishments: We have accomplished the following in this project.

a. An L-shape optical table has been acquired and installed. It provides a stable and isolated platform for the laser system.

b. An ultrafast amplifier laser system, as described in the proposal, has been acquired and installed. The purchase order for the laser system was placed in October 2017. The system was delivered in January 2018 and installed in February 2018. The laser system has been in operation.

c. A user training session was held in August 2018. Potential users from four different departments at FIU attended a user training session in August. The training included: 1) Introduction of the laser system and possible applications, 2) Daily operation of the laser system, 3) Q&A, and 4) Lab tour.

d. The laser system has been used by FIU researchers and students to perform experiments in several material research projects. Some examples of these projects are:
1) Two-dimensional Coherent Spectroscopy (2DCS) of monolayer transition metal dichalcogenides (TMDs). 2DCS has some unique advantages for studying valley dynamics in 2D TMD materials such as separating homogeneous and inhomogeneous linewidths, isolating relaxation pathways, and detecting valley coherence. However, performing 2DCS on monolayer TMDs is technically challenging due to short coherence time, monolayer thickness, and scattering from substrate. We addressed these technical difficulties and are able to consistently acquire reliable 2D spectra from monolayer TMDs. We obtained a series of 2D spectra for the trion resonance in monolayer MoSe2 at different excitation densities and sample temperatures. The homogeneous linewidth can be extracted by fitting the cross-diagonal linewidth in 2D spectra. The results show that the homogeneous linewidth increases linearly with the excitation density due to excitation induced dephasing. The linewidth at zero excitation density can be extrapolated from the excitation dependence. This measurement was repeated at various temperatures to extrapolate the residue homogeneous linewidth at zero excitation and zero temperature, which gives an intrinsic coherence time of 182 fs. By using proper combination of excitation pulse polarizations, 2DCS can selectively excite and detect a particular valley polarization and coherence. We have obtained 2D spectra with various combination of polarizations that provide direct evidence for valley exchange and valley coherence and also allow studies of their dynamics. This work has been published in Physical Review Materials 2, 054001 (2018).

2) Ultrafast dynamical study of carriers in Perovskites. Metal halide perovskite has become an attractive material

for solar cell application due to rapid increase in efficiency. Due to its tunability in optoelectronic properties and easy fabrication, its application has expanded to light-emitting diodes, lasers, photodetectors, spintronics, etc. Methylammonium lead iodide (MAPbI3) is the most commonly used perovskite material. We performed 2DCS on a MAPbI3 film at 115 K. At this temperature, there are two PL peaks corresponding to two resonances of different phases. We studied the ultrafast dynamics of the two phases as well as the coupling between the two phases. The energy transfer process was observed between the two phases. We also found that coherent phonon might play a role in the coupling of the two phases. The work has been summarized in a manuscript that is to be submitted soon.

Training Opportunities: A training session of the potential users was held in August. Potential users from four different departments (Physics, Chemistry, Electrical Engineering, and Material Engineering) attended the training session. The training included: 1) Introduction of the laser system and possible applications, 2) Daily operation of the laser system, 3) Q&A, and 4) Lab tour.

Results Dissemination: Publications:

1. Y. Hu, F. Zhang, M. Titze, B. Deng, H. Li*, and G.J. Cheng*, “Straining effects in MoS2 monolayer on nanostructured substrates: temperature dependent photoluminescence and exciton dynamics,” Nanoscale 10, 5717 (2018).

2. M. Titze, B. Li, X. Zhang, P.M. Ajayan, and H. Li, “Intrinsic coherence time of trions in monolayer MoSe2 measured via two-dimensional coherent spectroscopy,” Physical Review Materials 2, 054001 (2018).

3. M. Titze, C. Fei, M. Munoz, H. Wang, and H. Li, "Carrier Dynamics between the Ordered and Disordered Orthorhombic Phase in Methylammonium Lead Iodide Perovskites Revealed by Two-Dimensional Coherent Spectroscopy," in preparation.

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Technology Transfer: Nothing to Report

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Article Title: Straining effects in MoS2 monolayer on nanostructured substrates: temperature-dependent photoluminescence and exciton dynamics
Authors: Yaowu Hu, Feng Zhang, Michael Titze, Biwei Deng, Hebin Li, Gary J. Cheng
Keywords: 2d materials, straining effects, ultrafast spectroscopy
Abstract: Strain-engineering of two-dimensional (2D) transition metal dichalcogenides (TMDs) has great potential to alter their electronic and optical properties. Thus far, experimental studies of the straining effects in 2DTMDs primarily focused on the static property measurements at room temperature. However, low-temperature and temperature-dependence studies are essential in understanding the underlying mechanisms of the unique properties of monolayer TMDs. Herein, the temperature-dependent dynamic properties of laser shock strain-engineered monolayer MoS2 were studied using temperature-dependent photoluminescence (PL) and pump–probe spectroscopy. Both the photoluminescence spectra and exciton dynamics exhibit the differences between the MoS2 monolayers transferred on the flat and nanostructured surfaces by laser shock strain engineering and display a strong temperature dependence. The laser-induced straining effect and temperature-dependent dynamic behavior of MoS2 were studied through molec
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Authors: Michael Titze, Bo Li, Xiang Zhang, Pulickel M. Ajayan, Hebin Li
Keywords: Excitons, Trions, Valleytronics, Transition-metal dichalcogenide, Femtosecond laser spectroscopy, Four-wave mixing
Abstract: Quantum coherence and its dynamics in monolayer transition metal dichalcogenides (TMDs) are essential information to fully control valley pseudospin for valleytronics applications. Experimental understanding of coherence dephasing dynamics has been limited for excitons and largely unexplored for trions in monolayer TMDs. Here we use optical two-dimensional coherent spectroscopy to measure the trion coherence dephasing time in monolayer MoSe2 by analyzing the homogeneous linewidth. An intrinsic coherence time of 182 fs is extrapolated from the excitation density and temperature dependence measurement. The results show that trion-trion and trion-phonon interactions strongly affect the coherence dephasing time, while the intrinsic coherence time at zero excitation and zero temperature is primarily limited by the pure dephasing due to defect states. Our experiment also confirms optical two-dimensional coherent spectroscopy as a reliable technique for studying valley quantum dynamics in two
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Nothing to report in the uploaded pdf (see accomplishments)