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BSc, (Physics) Kings College and Dalhousie University, 2009
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DEPARTMENT OF PHYSICS AND ATMOSPHERIC SCIENCE

TITLE OF THESIS:	FOUR-WAVE MIXING EXPERIMENTS ON SOLUTION-PROCESSED METHYLAMMONIUM LEAD IODIDE (CH3NH3PBI3) PEROVSKITE THIN FILMS
TIME/DATE:	9:00 am, Friday, December 7, 2018
PLACE:	Room 1020, Kenneth C. Rowe Management Building, 6100 University Avenue

EXAMINING COMMITTEE:

Dr. Carlos Silva, School of Physics, Georgia Institute of Technology (External Examiner)

Dr. Ian Hill, Department of Physics and Atmospheric Science, Dalhousie University (Reader)

Dr. Laurent Kreplak, Department of Physics and Atmospheric Science, Dalhousie University (Reader)

Dr. Kimberley Hall, Department of Physics and Atmospheric Science, Dalhousie University (Supervisor)

DEPARTMENTAL REPRESENTATIVE:	Dr. Andrew Rutenberg, Department of Physics and Atmospheric Science, Dalhousie University
CHAIR:	Dr. Norman Schepp, PhD Defence Panel, Faculty of Graduate Studies

ABSTRACT

Hybrid organic-inorganic perovskite materials have gained widespread interest in recent years due to the high solar cell efficiencies demonstrated using perovskite as the absorbing layer. These materials may be deposited with a low-cost solution processing technique, yet solar cell efficiencies as high as the commercial standard(made from Silicon, grown using an expensive, high temperature process) have been achieved. The surprising performance of perovskite solar cells leads to many open questions regarding the optoelectronic properties of these interesting materials. This thesis work aims to shed light on these optoelectronic properties of the archetypical perovskite material CH3NH3PbI3 (MAPI) by applying the coherent optical technique of femtosecond four-wave mixing (FWM). In contrast to incoherent optical techniques such as transient absorption and photoluminescence, FWM probes coherence excited on the electron-hole pairs in the semiconductor, opening new opportunities to study the fundamental photo species and scattering processes.

We used FWM to directly determine the free exciton binding energy as well as the binding energy of excitons bound to shallow trap states in the MAPI system. Using FWM we were able to clearly decipher the exciton signal from the free-carrier continuum response, not apparent in incoherent spectroscopy signals due to the broadening associated with the soft nature of the organic-inorganic perovskite lattice. FWM was also used to measure the carrier-carrier scattering rate in MAPI thin-films, and was compared to GaAs. It was found that carrier-carrier scattering is much weaker in the MAPI system compared to GaAs over the carrier densities probed reflecting the operating densities in solar cells, resulting in a stark contrast between hybrid MAPI and the archetypical semiconductor GaAs. Finally, we used FWM to measure the dephasing time as a function of temperature, and as a function of excess energy near the bandgap. The results fit a model of electron-phonon scattering that included contributions from impurity scattering, and scattering by both acoustic phonons and optical phonons. These results show that for MAPI the recently-discovered Rashba effect enhances the rate of acoustic phonon scattering.