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ABSTRACT

Understanding the mechanisms responsible for the relaxation of charge carriers in semiconductor systems is crucial for the development of novel devices based on these materials. Transient four-wave mixing (FWM) is a powerful technique used to study relaxation processes as it is intimately connected to these mechanisms. In this thesis work, three different experimental implementations of FWM were used to study three different semiconductor systems relevant for optoelectronic applications. FWM was used to probe charge diffusion in CH₃NH₃PbI₃. CH₃NH₃PbI₃ is an attractive material for solar cell devices due in part to its large charge diffusion length. In this work, charge transport in CH₃NH₃PbI₃ was directly measured resulting in a calculated ambipolar diffusion length of 0.95 μm . Relative to the measured grain size in this sample, the larger diffusion length suggests that grain boundaries do not significantly impact charge transport.

The properties of GaAs grown at lower than conventional temperatures can be tailored via post-growth annealing. Spectrally-resolved FWM (SR-FWM) was used to study the effect of annealing on the coherent response of LT-GaAs. For low annealing temperatures, an observed dip in the SR-FWM response was found to stem from a polarization interference between the many-body exciton response and that of the band tail response. The interband dephasing time was observed to increase for increasing annealing temperatures. Extracted values for the Urbach energy of band tail states revealed a dramatic decrease at 550°C.

SR-FWM and two-dimensional Fourier transform spectroscopy (2DFTS) were used to study the interactions between bound and unbound electron-hole pairs in GaAs. Through comparison with numerical simulations of the 2DFTS response it was determined that exciton-carrier scattering was ten-fold stronger than exciton-exciton scattering, and that excitation-induced dephasing manifested in the real part of the 2DFTS spectra stronger than excitation-induced shift.

DEPARTMENT OF PHYSICS AND ATMOSPHERIC SCIENCE

TITLE OF THESIS: TRANSIENT FOUR-WAVE MIXING STUDIES OF GaAs, LOW-TEMPERATURE-GROWN GaAs, AND CH₃NH₃PbI₃

TIME/DATE: 1:30 pm, Thursday, December 7, 2017

PLACE: Room 3107, The Mona Campbell Building, 1459 LeMarchant Street

EXAMINING COMMITTEE:

Dr. Mark R. Freeman, Department of Physics, University of Alberta
(External Examiner)

Dr. Kevin Hewitt, Department of Physics and Atmospheric Science,
Dalhousie University (Reader)

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

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

DEPARTMENTAL REPRESENTATIVE: Dr. Harm Rotermund, Department of Physics and Atmospheric Science, Dalhousie University

CHAIR: Dr. Zhizhang Chen, PhD Defence Panel, Faculty of Graduate Studies