Ultrafast Spectroscopic Probes of Acoustic Phonons in Multiferroic Thin Films and Semiconductor Heterostructures

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***Abstract* — The interaction of an ultrashort pulse of light with acoustic phonons provides a convenient and non-contact methodology to determine the elastic properties of nanomaterials. The epitaxial growth of thin films with thicknesses of a few hundred nanometres can create multiferroics with properties tailored by strain engineering. Ultrafast photoacoustic spectroscopy was used to determine the elastic constant and magnon response of the room temperature magnetoelectric bismuth ferrite. When applied to semiconductor quantum well heterostructures photoacoustic spectroscopy provides a precise probe of superlattice periodicity and interface quality.**

***Index Terms* — acoustic phonons; multiferroics; quantum cascade lasers; thin films; ultrafast spectroscopy.**

I. INTRODUCTION

Traditional methods by which the acoustic properties of bulk materials are determined are reliant upon the use of a piezoelectric transducer to create and sense an acoustic pulse. However, round-trip times for materials with sub-micron thickness are typically 100ps, faster than achievable with conventional electronics.

Time-resolved reflectivity using femtosecond pump and probe pulses provides a convenient, non-contact means to determine the acoustic phonon velocity and dispersion in nanomaterials. Below two examples of our recent work using picosecond ultrasonic spectroscopy are highlighted.

II. MULTIFERROIC THIN FILMS

Multiferroic materials exhibit multiple hysteretic orders, such as ferromagnetism and ferroelectricity [1]. In magnetoelectric multiferroics these properties are intimately coupled - allowing, for instance, the electrical switching of magnetisation. Bismuth ferrite (BFO) is a room temperature multiferroic, and exhibits both weak ferromagnetism and ferroelectricity in thin-film form [2].

Using two-colour time-resolved reflectivity we created acoustic phonon pulses (400nm pump) and tracked their propagation (800nm probe) through BFO thin films and into the substrate. Low frequency (<40GHz) modes were assigned to longituidinal acoustic phonons (see Figure), while pronounced magnon modes were observed in Dy-doped and Dy and La co-doped.

III. QUANTUM CASCADE STRUCTURES

Quantum cascade lasers (QCLs) are compact and convenient electrically-pumped sources of light in the mid- and far-infrared. Terahertz-frequency QCLs [3] are currently limited to cryogenic temperatures, and are reliant upon precise epitaxial growth of high quality quantum well interfaces. We have used ultrafast time-resolved reflectivity to investigate the acoustic phonon dispersion of THz-QCL heterostructures fabricated from GaAs quantum wells and AlGaAs barriers [4].

Zone-folding of the acoustic phonons created numerous optically-active modes that enabled superlattice periodicities to be determined with a precision of 0.1%. Rougher interfaces were found to degrade the number of zone-folded modes seen at higher frequencies. The impulsive resonant generation of the acoustic phonons was studied by wavelength and temperature tuning [4].

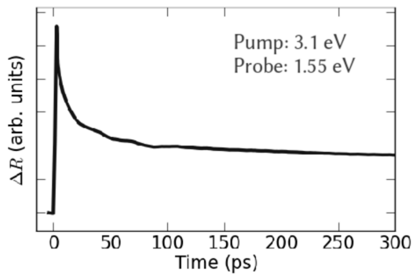

Figure - Oscillations in the time resolved reflectivity of a thin film of the multiferroic bismuth ferrite from acoustic phonons in the film (<100ps) and substrate (>100ps).

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