iJRASET
International Journal For Research in Applied Science and Engineering Technology

INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

Volume: 6 **Issue:** II **Month of publication:** February 2018

DOI:

www.ijraset.com

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International Journal for Research in Applied Science & Engineering Technology (IJRASET)
ISSN: 2321-9653; IC Value: 45.98; SJ Impact Factor: 6.887
Volume 6 Issue II, February 2018- Available at www.ijraset.com

Terahertz Time-Domain Spectroscopy of Nitrogen Ice

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Abstract: Terahertz time-domain spectroscopy of Nitrogen ice is performed between 10 K and 30 K. The optical rectification is utilized to generate the terahertz radiation and electro-optic sampling is utilized to detect the radiation. The sample was grown in a cryostat via vapor deposition. The Nitrogen ice has absorption feature around 1.45 THz that changes with temperature.
Keywords: Nitrogen Ice, Terahertz, Vapor deposition

I. INTRODUCTION

Nitrogen (N_2) liquifies at 77 K and solidifies at 63.1 K. For temperatures between 63.1 K and 35.6 K, it takes hcp crystal structure and D_{46h} space group (β-phase) and for temperatures less than 35.6 K, it takes fcc crystal structure and T_{6h} space group (α phase) [1]. In the α-phase, N_2 ice has two triply degenerate infrared active phonons of T_u symmetry and two T_g and one E_g symmetric librons [2,3]. The lattice vibrational frequencies of α-N_2 were calculated by assuming various intermolecular pair potentials [2,4]. Among all the potentials, the one written as sum of Lennard-Jones and quadruple-quadrupole interaction terms is the best to calculate the phonons of α-N_2 ice. We grew the Nitrogen ice by vapor deposition technique and studied it by our terahertz time domain spectroscopy setup. We have observed shifting of the center frequency of the absorption line in Nitrogen around 1.45 THz to smaller values as the temperature is increased.

II. EXPERIMENTAL SETUP

Figure1 shows the schematic of the experimental setup. We built a terahertz time-domain spectrometer and integrated that with a cryostat in which we grew N_2 ice. The Terahertz radiation (THz) was generated by optical rectification inside a (110) oriented 1mm thick ZnTe crystal. The THz was guided by a couple of off axis parabolic mirrors to a frequency independent spot at which a sapphire substrate attached to the cold finger of the cryostat is located.

The transmitted THz through the substrate is guided by another pair of identical off axis parabolic mirrors onto another (110) oriented 1mm thick ZnTe crystal for the detection of THz by electro-optic sampling. The entire experiment was enclosed in a box and purged with dry Nitrogen gas to bring the water vapor percent down. A hygrometer located in the experiment box measures the relative humidity in the box. The temperature controlling and the acquisition of the data of relative humidity in the box and the THz waveform was done by a Lab View program.

The cryostat has two sapphire windows for passing the THz radiation through it. Through the edge of the front window of the cryostat, a steel gas line is drawn into the cryostat that faces the substrate normally. The other end of the steel line is attached to a N_2 gas bulb at 300 torr via a flow control valve. We evacuated the cryostat up to 4μtorr pressure before cooling the substrate. We took a reference THz waveform at 10 K transmitted through the bare substrate. When the N_2 gas valve is released, the solid N_2 is grown on the substrate at 10 K by the deposition of the gas.

The deposition was done for 2 hrs and the shift of sample waveform peak from the reference peak suggests a rate of deposition of 370 μm per hour. We acquired THz waveforms transmitted through the N_2 ice maintained at 10K, 15 K, 20 K, 22 K, 25 K, 27K, and 30 K. We calculated the Fourier transformations of the waveforms using our MATLAB code. Since we analyzed the waveforms of 22 ps length, our spectral resolution is 45.45 GHz. The relative humidity of the experiment box was maintained at less than 1% RH throughout the experiment.

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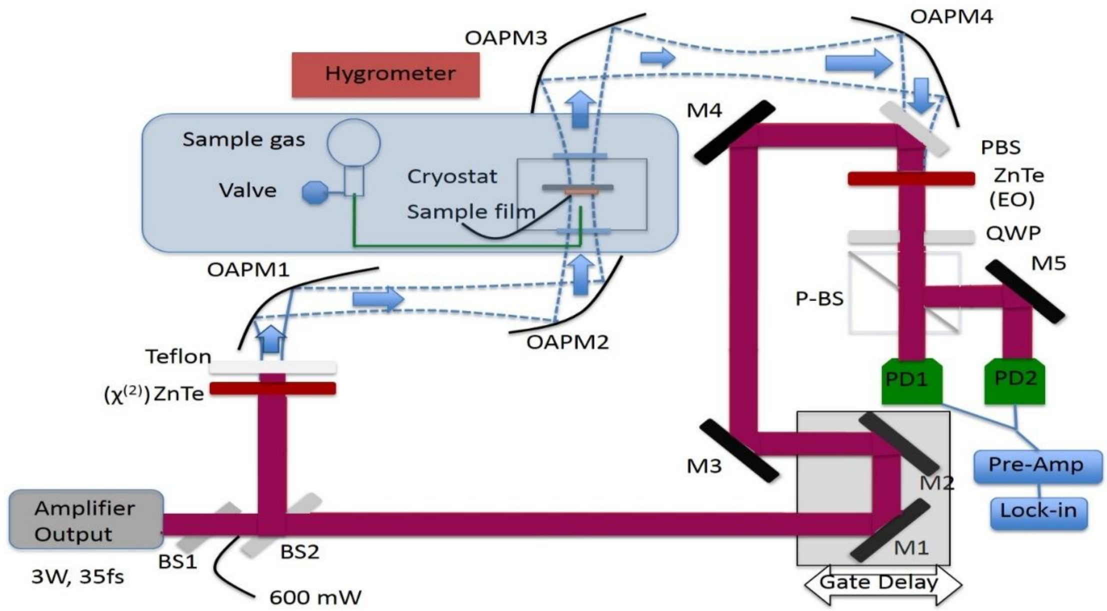

Figure 1: Experimental setup to grow thin ice films and study them with THz-TDS. Here, BS1: 80% reflecting beam splitter, BS2: 95% reflecting beam splitter, OAPM: Off-Axis Parabolic Mirror, M1-M5: Mirrors for 800 nm wavelength, PBS: Pellicle beam splitter, PBS: Polarization beam splitter, QWP: Quarter wave plate, PD1, PD2: Silicon Photodiodes, Solid black lines (red in electronic version): Optical beam, Grey dashed curves (light blue in electronic version): The boundary of THz radiation, and Arrows: Propagation direction of THz radiation. Pre-Amp: Current pre-amplifier. Lock-in: Lock-in amplifier.

III. RESULTS

The transmission co-efficient is defined as the ratio of Fourier transformations of waveforms transmitted through the sample and the reference. Figure 2 (top) shows the THz waveforms transmitted through reference and nitrogen ice at different temperature. Figure 2 (bottom) shows the amplitude of the transmission coefficient as a function of temperature of N_2 ice. In the amplitude spectrum of transmission co-efficient, we can see that the center frequency of absorption around 1.45 THz is decreasing to the smaller frequencies when the temperature of the ice is increased. Similar trend in the data was observed even in the high-resolution far-infrared spectroscopy of Nitrogen ice [7]. Since the observed absorption center frequencies are close to the values predicted by the phonon theories [5], the absorption may be due to coupling of THz photons to the lower frequency T_u phonons of Nitrogen ice.

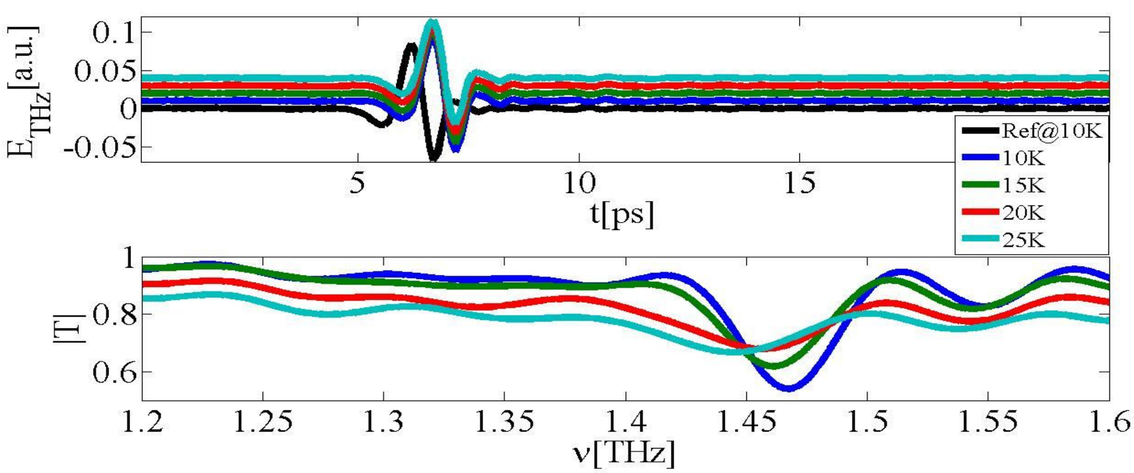

Figure2: (top) Terahertz waveforms transmitted through the reference and Nitrogen ice at temperature 10 K, 15 K, 20 K, and 25 K. The waveforms are shifted up just clarity in the figure. (bottom) Terahertz amplitude transmission coefficient of Nitrogen ice at 10 K, 15 K, 20 K, and 25 K.

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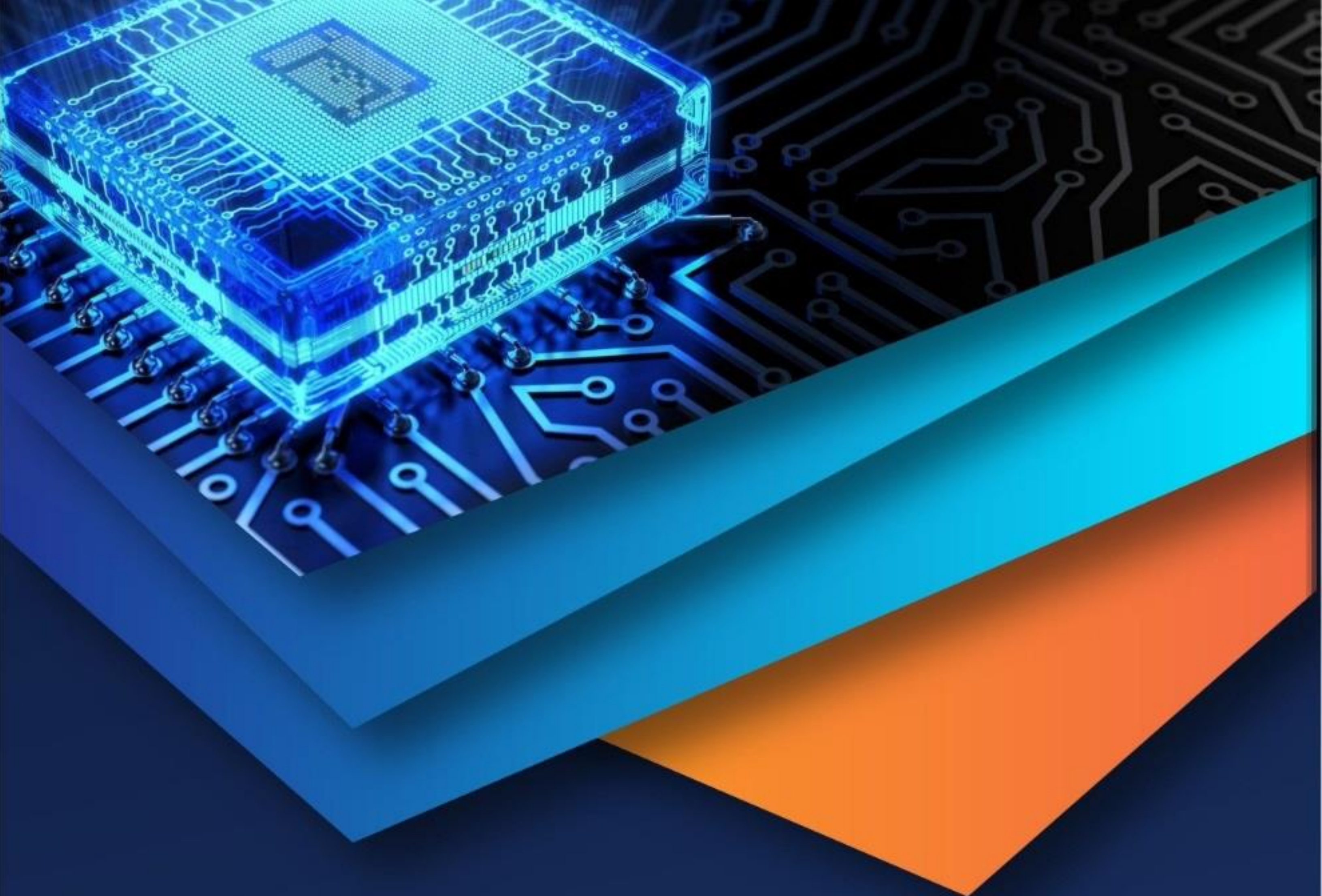

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