ULTRASONIC SENSING OF THE LOCATION AND SHAPE OF A SOLID/LIQUID INTERFACE FOR CRYSTAL GROWTH CONTROL

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INTRODUCTION

Modern infrared focal plane arrays consist of Hg$_{1-x}$Cd$_x$Te deposited on lattice matched Cd$_{1-x}$Zn$_x$Te substrate wafers. The performance of these infrared focal plane arrays is, in large part, dependent on the quality of the substrate. Cd$_{1-x}$Zn$_x$Te substrates are machined from bulk crystals, grown using a vertical Bridgman technique and their quality is governed by the conditions of solidification. In the vertical Bridgman method, the bulk charge is first melted in a cylindrical quartz ampoule, and then slowly drawn from the hot zone (~1100°C) through a gradient section into a cold zone (~750°C) [1]. For all single crystal materials grown by this technique, the highest quality crystal is achieved with a planar or near planar (slightly convex towards liquid) interface shape during growth and a small thermal gradient during cooling [2]. There is a need for a sensor technology that can measure these parameters in real time throughout the growth process.

Ultrasonic sensing by the pulse-echo method has been used to monitor the location and shape of the solid/liquid interface during Bridgman growth [3,4]. This method is based on the observation that there is an acoustic impedance difference between the solid and liquid phases and this difference causes an acoustic reflection at the solid/liquid interface. However, this echo signal may be weak and difficult to detect. Through transmission time of flight (TOF) measurements are less susceptible to this, and have been used to locate the solid/liquid interface in the solidification of aluminum alloys [5]. This method is based on the observation that an abrupt drop (5-20%) in ultrasonic velocity occurs as the solid transforms to the liquid phase. We are investigating the ultrasonic TOF measurements for in-situ monitoring of the shape and location of the solid/liquid interface during vertical Bridgman growth of Cd$_{1-x}$Zn$_x$Te.
Figure 1 Bench top model to simulate the curvature of the solidifying interface.

To optimize the application of this method to single crystal growth, we have designed a bench-top model to simulate the solidifying interface during growth. The model system uses polymethylmethacrylate (PMMA) as the solid phase and water as the liquid phase (Figure 1). These materials were chosen because they have similar ultrasonic velocities to those of solid and liquid Cd$_{1-x}$Zn$_x$Te, respectively. We have (1) designed a laser ultrasonic sensor to measure the TOF of an acoustic wave in the sagittal plane for a range of interface heights, (2) developed a 2-D ray tracing algorithm to predict the minimum TOF (or first arrival) of the propagating wave front, and (3) used a nonlinear least squares algorithm based on the ray tracing model to reconstruct the interface radius and velocity values of the solid and liquid phases. Encouraging results have been obtained.

METHOD AND APPARATUS

The laser ultrasonic sensor used to evaluate the model system is shown in Figure 2. The acoustic wave is generated by a pulsed Nd:YAG laser delivering 10ns duration pulses of 50mJ energy. The laser beam is focused to a circular spot size of 1mm diameter producing an instantaneous power density of ~60 MW/cm$^2$, which is slightly ablative [6]. A glass surface constraint with propylene glycol as a couplant was used to further enhance signal generation. The laser generated wave was propagated through the sample and detected at the epicenter with a heterodyne Mach-Zehnder type interferometer [7].
This interferometer has a bandwidth of 1 KHz to 35 MHz, however to reduce noise, the signal was filtered from 10 KHz to 10 MHz. It uses an externally coupled 1 W Argon laser. The TOF signals were recorded at a sampling rate of 5ns, stored on a precision digital oscilloscope and downloaded to a computer for analysis. Each signal was the average of 100 shots to increase the signal to noise ratio of the waveform.

A schematic of the bench top model samples used in these experiments is shown in Figure 1. PMMA cylinders with known interface heights \( h \) of 0.0, 5.0, 10.0, and 15.0 mm were machined and placed in an aluminum tube. The tubes were then filled with water. TOF waveforms were acquired along the diameter of the sagittal plane as the laser source and receiver were vertically scanned from a position 10 mm below the interface (i.e. in the pure solid phase) through the interface region to a position 10 mm above the interface (i.e. in the pure liquid phase). The longitudinal wave velocity of PMMA is 2.67 mm/\( \mu \)s (for Cd\(_{1-x}\)Zn\(_x\)Te it can vary from 3.02-3.44 mm/\( \mu \)s at 25°C depending on the crystal orientation) [8]. The water had a velocity of 1.497 mm/\( \mu \)s (the velocity of liquid Cd\(_{1-x}\)Zn\(_x\)Te is unknown.

Although the source and receiver points were diametrically located, the ray path of the first arrival was not always a straight line due to ray refraction at the interface. The refraction of an ultrasonic ray as it passes through a solid/liquid interface is determined by Snell's law:

\[
\frac{\sin \theta_s}{V_s} = \frac{\sin \theta_l}{V_l}
\]
Figure 3 Ultrasonic ray paths for rays traveling in the liquid only region, interface region and the solid only region.

where \( V_s \) is the longitudinal wave velocity for the solid phase, \( V_l \) is the velocity for the liquid phase and \( \theta_s \) and \( \theta_l \) are the incident and transmitted angles of the wave with respect to the surface normal.

Examples of predicted ultrasonic ray paths for the model system are shown in Figure 3. The TOF for a wave propagating in only the liquid phase
\[
\text{TOF}_l = \frac{(x_r - x_s)}{V_l}
\]
which is simply the straight line propagation distance between the source \((x_s)\) and receiver \((x_r)\) positions, divided by the velocity of the liquid. Similarly the TOF for a wave propagating in the solid only phase
\[
\text{TOF}_s = \frac{(x_r - x_s)}{V_s}
\]
which is the same distance the ray propagated in the liquid but divided by the longitudinal wave velocity of the solid. Because of the velocity difference between the liquid and solid phases, \(\text{TOF}_l\) is \(~50\mu s\) whilst \(\text{TOF}_s\) is \(~28\mu s\).

The TOF for a wave propagating in the interface region depends on the fraction of the ray traveling in the liquid and solid phases. For a wave to be generated and detected in the same cross sectional plane, it must travel along a path in the solid phase that is parallel to the source/receiver plane as shown in Figure 3. This is because when the ray passes through the liquid/solid interface its angle of refraction is the same as when it passes through the solid/liquid interface. Therefore, for a ray generated and detected at the same axial position, the fraction of the ray propagating in the solid must be parallel to the source/receiver plane. The TOF for a ray propagating in this region is
\[ \text{TOF}_1 = \frac{2 \sqrt{(x_s - x_1)^2 + (z_s - z_1)^2}}{V_1} + \frac{x_2 - x_1}{V_s} \]

where \((x_1, z_1)\) and \((x_2, z_2)\) are the coordinates of ray path intersection at the interface (Figure 3). Due to the symmetry argument above, the distance the ray propagates in the liquid phase is the same on the source and receiver sides.

As the source and receiver are axially translated from the solid only phase through the interface, the distance propagated in the liquid increases and that in the solid decreases. Accordingly, the TOF in the interface region increases as the source/receiver plane is moved towards the liquid phase. This effect can be seen in the expanded section of Figure 3. From this 2-D ray tracing code we were able to predict the first arrival TOF as a function of position along the axial diameter of the sagittal plane.

RESULTS AND RECONSTRUCTION

Figures 4, 5 and 6 show both the experimental and predicted TOFs (from the 2-D ray tracing code) for \(h\) values of 0.0, 5.0 and 10.0mm respectively. Figures 5 and 6 show a gradual increase in the TOF as the source/receiver is translated from the solid only phase into the interface region. The tip of the interface is noted by an abrupt shift in the TOF to the value for a ray propagating in the liquid only phase. For a planar interface (Figure 4), there is only an abrupt increase in TOF at the solid/liquid interface from \(\sim 28\mu s\) to \(\sim 50\mu s\).

For non-planar interfaces, we only need to recover the interface radius and the velocity values of the liquid and solid phase. Once these values are known, the interface height and error between true and reconstructed values were calculated. We have used a nonlinear least squares method that utilizes predictions from the 2-D ray tracing code to reconstruct the interface radius and velocity values of the liquid and solid phase [9]. Table I shows the results of the nonlinear least squares reconstruction for interface heights of 5.0, 10.0, and 15.0mm, using 15, 20 and 25 TOF values obtained by translating the source and receiver in increments of 1.0mm through the interface.

The nonlinear least squares reconstruction has recovered the interface radius and velocity values for the model reasonably well. Back calculating the interface height from the recovered values has resulted in determining \(h\) to within 0.3mm for interface heights of 5.0 and 10.0mm. For \(h\) of 15.0mm, we determined the interface height to be within 1.7mm. For the planar interface, the location was determined by the abrupt drop in ultrasonic TOF as the propagating ray was axially translated from the liquid only phase to the solid only phase. The location was determined to within 0.5mm, which was the step size in the axial translation. This level of reconstruction accuracy appears satisfactory for the needs of crystal growth, and studies are underway to make measurements during the growth process.
Figure 4  Interface mapping of the TOF for $h = 0.0$mm (i.e. a planar interface).

Table I

NONLINEAR LEAST SQUARES RECONSTRUCTION RESULTS

<table>
<thead>
<tr>
<th></th>
<th>Interface Height $h$ (mm)</th>
<th>Radius of Curvature (mm)</th>
<th>Water Velocity (mm/μs)</th>
<th>PMMA Velocity (mm/μs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>True Value</td>
<td>5.00</td>
<td>139.14</td>
<td>1.497</td>
<td>2.670</td>
</tr>
<tr>
<td>Calc. Value</td>
<td>4.91</td>
<td>141.56</td>
<td>1.398</td>
<td>2.671</td>
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<tr>
<td>Error</td>
<td>1.80%</td>
<td>1.74%</td>
<td>6.04%</td>
<td>0.05%</td>
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<tr>
<td>True Value</td>
<td>10.00</td>
<td>73.45</td>
<td>1.497</td>
<td>2.670</td>
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<tr>
<td>Calc. Value</td>
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<td>75.32</td>
<td>1.553</td>
<td>2.612</td>
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<tr>
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<td>2.54%</td>
<td>3.74%</td>
<td>2.16%</td>
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<tr>
<td>True Value</td>
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<td>53.07</td>
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<tr>
<td>Calc. Value</td>
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<td>57.89</td>
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<tr>
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<td>11.02%</td>
<td>9.08%</td>
<td>8.02%</td>
<td>0.09%</td>
</tr>
</tbody>
</table>
Figure 5 Interface mapping of the TOF for $h = 5.0 \text{mm}$.

Figure 6 Interface mapping of the TOF for $h = 10.0 \text{mm}$. 
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REFERENCES