SOLID-LIQUID INTERFACE RECONSTRUCTIONS USING LASER ULTRASONIC FAN-BEAM PROJECTION DATA

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INTRODUCTION

Many single crystal semiconductors are grown by variants of the Bridgman technique in which a cylindrical ampoule containing a molten semiconductor is translated through a thermal gradient, resulting in directional solidification and the growth of a single crystal. During crystal growth, the shape and location of the solid-liquid interface together with the local temperature gradient control the mechanism of solidification (i.e. planar, cellular or dendritic), the likelihood of secondary grain nucleation/twin formation (i.e. loss of single crystallinity), solute (dopant) segregation, dislocation generation, etc. and thus determine the crystals’ quality [1]. For crystals grown by the vertical Bridgman (VB) technique, optimum properties are obtained with a low (~1.5mm/hr) constant solidification velocity and a planar or near planar (slightly convex towards liquid) interface shape maintained throughout growth [2,3]. The solidification rate and the interface shape are both sensitive functions of the internal temperature gradient (both axial and radial) during solidification, which is governed by the heat flux distribution incident upon the ampoule, the latent heat release at the interface, and heat transport (by a combination of conduction, buoyancy surface tension driven convection and radiation) within the ampoule [4,5]. The solid-liquid interface’s instantaneous location, velocity and shape during crystal growth are therefore difficult to predict and to control, especially for those semiconductor materials with low thermal conductivity (i.e. CdZnTe alloys) [6]. Thus the development of ultrasonic technologies to non-invasively sense the interface location and shape throughout VB crystal growth processes has become a key step in developing a better understanding of the growth process and for enabling eventual sensor-based manufacturing.

Here, we experimentally explore the use of a laser ultrasonic sensor concept for monitoring the solid-liquid interface location and shape. A combination of ray path analysis and testing of a model (isotropic) system (where interface geometries are precisely known) were used to evaluate sensing and interface reconstruction approaches. While semiconductor crystals are elastically anisotropic, modeling has suggested that strategies that work on isotropic systems can be readily extrapolated to anisotropic ones, provided point sources and receivers are used, and an anisotropic generalization of Snell’s law is incorporated in the ray tracing [7].
MODEL SYSTEM

The bench-top model with known, solid-liquid velocities, consisted of water and solid PMMA contained in a cylindrical aluminum (2024-T6) "ampoule" because opaque ampoules such as pyrolitic boron nitride (PBN) are sometimes used during crystal growth. By machining the end of the PMMA, the interfacial curvature could be varied from convex, planar to concave (viewed toward the liquid). The interface convexities h (defined such that h > 0 corresponds to a convex interface) studied were h = ±2, ±5, ±10mm and planar (h = 0mm), Figure 1. The water and PMMA had a measured longitudinal wave velocity of 1.50 ± 0.01 mm/μs and 2.67 ± 0.01mm/μs at 21°C, respectively. While the 2024-T6 Al alloy had a measured longitudinal wave velocity of 6.35 ± 0.01mm/μs and a shear wave velocity of 3.01 ± 0.01mm/μs at 21°C.

LASER ULTRASONIC MEASUREMENTS

The ultrasonic time-of-flight (TOF) between precisely located source and receiver points was measured using the laser ultrasonic system shown in Figure 2. A ~10ns duration Q-switched Nd:YAG laser pulse of 1.064-μm wavelength was used as the ultrasonic source. The energy per pulse was ~15mJ and the roughly Gaussian beam of the multimode pulse was focused to an approximate circular spot 1mm in diameter. Thus, the source power density was ~750MW/cm². The low infrared absorption coefficient for the aluminum required the use of a constraining layer consisting of a glass slide and a propylene glycol couplant. The ultrasonic receiver was a heterodyne laser interferometer, which responded to the sample's out-of-plane (normal) surface displacement associated with wavefront arrivals at the receiver point. It was powered by a 1-W single mode argon ion laser (operated at 0.25W), which produced a continuous Gaussian beam of 514-nm wavelength focused to a circular spot on the sample ~100μm in diameter. The signal from the interferometer was bandpass filtered between 10kHz and 10MHz and recorded with a precision digital oscilloscope at a 2ns sampling interval using 8-bit analog-to-digital conversion. To improve the signal to noise ratio, each waveform used for a TOF measurement was the average of ~25 pulses collected at a pulse repetition rate of 20Hz. A fast photodiode identified the origination time for the ultrasonic signals.

![Diagram](image.png)

Figure 1. A schematic of the bench-top model showing the solid-liquid interface and the velocity fields of the liquid and solid. (All dimensions in mm)
Eight ultrasonic TOF scans along a vertical diametral plane were conducted for each interface such that the receiver was located in both the liquid \((z_r = 5, 10, 15, \text{ and } 20\text{mm})\) and solid \((z_r = -5, -10, -15, \text{ and } -20\text{mm})\) while the source was axially scanned from the liquid phase \((z_s = 40\text{mm})\) to the solid phase \((z_s = -20\text{mm})\), resulting in a fan beam projection array. Errors in measured sample sizes, imprecision in the translation stage alignment, temperature fluctuations, etc., resulted in an estimated TOF error of about ±100ns.

RAY PATH AND TIME-OF-FLIGHT PREDICTIONS

Suppose an ultrasonic ray is incident upon an interface; both reflected and refracted rays propagate on the diametral plane (defined by the incident propagation vector and the normal to the interface at the intersection of the incident ray with the interface) [8]. One of the directions will result in a ray whose path usually reaches the receiver point. The time-of-flight, \(\tau_m\), is the integral of the inverse velocity field (slowness) for a ray of path length, \(L_m\), and is represented by

\[
\tau_m = \int_{L_m}^{\frac{dl}{v(x)}}, \quad m = 1, 2, \ldots M
\]

where \(dl\) is an infinitesimal element of the path, \(1/v(x)\) is the local slowness within the object and \(M\) is the number of different rays.

If the coordinates of the source \((x_s, z_s)\) and receiver \((x_r, z_r)\) points on the diametral plane are prescribed, then determining the ray path between these two points constitutes a boundary-value problem. The solutions of boundary-value problems like this are usually preceded by solutions of initial value problems[9] in which initial ray angles at the source point are prescribed and the ray paths are obtained by solving for the refraction angles at the interface. After obtaining the ray paths for an arbitrary set of initial ray angles emanating from the source point, the ray path between the prescribed source/receiver points can be obtained using the shooting method [10]. In this approach, an initial ray direction is first arbitrarily chosen, and the distance between the receiver point and the intersection of the ray path with the outer boundary calculated. The procedure is then repeated using a slightly different initial ray angle until the distance is smaller than a prescribed tolerance \(\delta\). Here 
\[
\delta = 2.5 \times 10^{-3} R_s
\]
was chosen, where \(R_s\) is the cylinder radius.
Calculated ray paths on a diametral plane for a convex interface \((h = 5\text{mm})\) are shown in Figures 3a and 3b for a source point located in the liquid \((z_s = 15\text{mm})\) and the solid \((z_s = -15\text{mm})\), respectively. When the source point is located in the liquid, Figure 3a, ray paths in the liquid are straight and are only bent during propagation through the interface with the solid. Note that some of the rays are refracted at the first liquid-solid interface travel through the solid and are again refracted at the second solid-liquid interface (doubly refracted rays) while others are only refracted once at the first liquid-solid interface (singly refracted ray). In this case, doubly refracted rays and rays with straight paths may both reach the same boundary point. However, these two kinds of rays are experimentally distinguishable because the doubly refracted ray has a shorter travel time and would have suffered an energy loss due to reflection and mode conversion each time it crossed the interface. The case of a source located in the solid for a convex interface \((h = 5\text{mm})\) is shown in Figure 3b. Here the case is much simpler, ray paths are either straight (in the solid) or are refracted only once by the interface (singly refracted ray).

Wavefronts separate ultrasonically disturbed regions from those that are undisturbed. Thus the TOF of an ultrasonic signal can in principle be measured at any receiver point on the sample’s periphery that is intersected by a wavefront. The wavefronts at any time after source excitation can be obtained by connecting points along the ray paths with the same travel time. On the diametral plane the calculated longitudinal wavefronts at 10µs intervals are also shown in Figures 3a and 3b. Ray paths and wavefronts for planar and concave interfaces \((h \leq 0)\) were calculated, however they are not shown here.

**TIME-OF-FLIGHT PROJECTIONS**

Measured TOF projections for convex interfaces \((h = 2, 5 \text{ and } 10 \text{ mm})\) are shown in Figures 4a and 4b for a receiver point located in either the liquid \((z_t = 15\text{mm})\) or the solid \((z_t = -15\text{mm})\) respectively. For the experiments it was more conducive to fix the receiver point \((z_t)\) and scan the source point \((z_s)\). For the case where the receiver is located in the liquid, Figure 4a, and the source also in the liquid \((z_s > 0)\) two wavefront arrivals corresponding to doubly refracted and non refracted rays were observed. From Figure 4a it is clear that for most sensor arrangements the TOF of doubly refracted rays is always smaller than those of non refracted (straight) rays and an energy loss was observed in the ultrasonic waveforms acquired.

![Figure 3](image.png)

*Figure 3. Ray paths and wavefronts on a diametral plane of a convex interface with \(h = 5\text{mm}\) for a source located in (a) the liquid \((z_s = 15\text{mm})\) and (b) the solid \((z_s = -15\text{mm})\).*
Therefore, these two arrivals were easily distinguishable. Now as the source was scanned into the solid \( z_s < 0 \) only one wavefront arrival was observed, corresponding to singly refracted rays. Now consider the receiver located in the solid and the source in the liquid \( z_s > 0 \), Figure 4b, only one wavefront arrival was observed, corresponding to singly refracted rays. Again as the source was scanned into the solid \( z_s < 0 \), only one wavefront arrival was observed, corresponding to non refracted (straight) rays. The measured TOF is in good agreement with that predicted by the refracted ray path models.

From direct inspection of the TOF projection data for convex interfaces it is clear that the interface location \( z_i \) can be easily identified by the abrupt change in ray propagation modes near the actual interface location \( z_i = 0 \). When the source was located in the liquid, there is a transition of doubly refracted and straight rays to singly refracted rays as the source was scanned from the liquid to the solid. Also the magnitude of the TOF traces of singly and doubly refracted rays decreased and there was a increase in the slope of doubly refracted rays as the interface convexity \( h \) increased. When the source was located in the solid, there was a transition of singly refracted rays to straight rays as the source was scanned from the liquid to the solid. Again the magnitude of the TOF traces of singly refracted rays decreased and slope increased as the interface convexity \( h \) increased. Therefore, the convexity of convex interfaces can only be qualitatively determined from direct observation of the TOF projection data, whereas the location \( z_i \) can be quantitatively evaluated. Similar results (not shown here) were observed with concave interfaces.

INTERFACE CURVATURE RECONSTRUCTIONS

In a crystal growth application, the location and curvature of the interface as well as the velocities of the solid/liquid regions are all unknown and must be determined from a set of TOF projection data. There are a variety of techniques available for reconstructing an object image from TOF projection data [11]. For crystal growth applications, approaches that can be used with sparse data and that exploits the often significant a priori information available are preferable. For example, direct inspection of the ultrasonic TOF projection data reveals that an interface does exist, experiments have shown that its shape can be approximated as a segment of a circle and for most situations, the gradient in temperature is small enough that the velocities are relatively uniform on either side of the interface. The use of a simple model of the solidification geometry with a small number of unknown parameters combined with the ray tracing analysis therefore enables the application of a least squares reconstruction approach.

![Image](image.png)

Figure 4. Ultrasonic time-of-flight projection data for convex interfaces, \( h = 2, 5 \) and 10mm, for a receiver located in (a) the liquid \( z_r = 15\text{mm} \) and (b) the solid \( z_r = -15\text{mm} \).
We assume the model geometry is of the form shown in Figure 1, where $h, z_i, v_l$ and $v_s$ are all unknown. For the refracted ray path model, the TOF depends nonlinearily on the interface convexity ($h$), interface location ($z_i$), liquid ($v_l$) and solid ($v_s$) velocities and the mean-square error is given by

$$
\chi^2 = \sum_{i=1}^{M} [\tau_i - \hat{\tau}_i(x_i; h, z_i, v_l, v_s)]^2
$$

where $\tau_i$ are the measured time-of-flights and $\hat{\tau}_i$ are the predicted time-of-flights for a model estimate of the interface. To reconstruct the model unknowns from the fan beam TOF projection data a Levenberg-Marquardt nonlinear least-squares reconstruction method was used [12]. The nonlinear least-squares algorithm returned the best-fit (i.e. reconstructed) parameters ($h, z_i, v_l, v_s$) by converging upon the interface that minimized $\chi^2$.

The reconstructed interface location ($z_i$) and convexity ($h$) obtained using the nonlinear least-squares method for convex interfaces with the receiver located in the liquid were within ±0.95mm and ±0.94mm, respectively for all interfaces ($h = 2, 5$ and 10mm) and receiver locations ($z_r = 5, 10, 15$ and 20mm). Also, the reconstructed liquid ($v_l$) and solid ($v_s$) velocities, were within 8.9% (for the liquid) and 5.3% (for the solid) of the actual velocities. Now for the receiver located in the solid, the interface location ($z_i$) and convexity ($h$) were within ±0.93mm and ±1.32mm, respectively for all interfaces ($h = 2, 5$ and 10mm) and receiver locations ($z_r = -5, -10, -15$ and -20mm). Again, the deduced liquid and solid velocities were within 4.9% (for the liquid) and 2.4% (for the solid) of the actual velocities. The reconstructed interfacial geometry of the convex interfaces for a receiver located in the liquid a) $z_r = 15$mm and the solid b) $z_r = -15$mm are shown in Figure 5a and b, respectively.

Crystal growth models are now successfully able to predict the general form of the location-time behavior and the form of interfacial curvature, but they do not reliably give the exact values. Using the general form of these solutions with free parameters ($h, z_i, v_l, v_s$) together with the nonlinear least-squares reconstruction routine appears to represent a robust approach for converging upon the correct interface model, and thus recovery of the interface geometry (i.e. solid-liquid interface location, convexity) and velocity fields from fan beam ultrasonic TOF data. Thus the approach promises to provide significant new information about the interface geometry to the crystal grower and may lead to a more detailed understanding of the growth process.

Figure 5. Nonlinear least-squares reconstructions for convex interfaces with the receiver located in a) the liquid ($z_r = 15$mm) and b) the solid ($z_r = -15$mm).
CONCLUSIONS

An experimental study of a potential laser ultrasonic sensor methodology for the sensing the solid-liquid interface location and shape similar to those encountered during vertical Bridgman growth of CdZnTe and other semiconducting materials has been conducted. A combination of ray path analysis and bench-top testing on a model isotropic solid-liquid interface with a prototype laser ultrasonic system was used to explore various sensing concepts and reconstruction methods to determine the solid-liquid interface location, interfacial curvature and resultant velocity fields from fan-beam ultrasonic TOF projection data collected on the diametral plane. From direct observation of the TOF projection data the interface location can be easily identified (by the discontinuity in ray propagation modes) whereas the convexity of an interface is only qualitatively determinable from the fan-beam TOF projection data. However, a nonlinear least-squares reconstruction routine robustly reconstructed the solid-liquid interface location, interface shape and local velocity fields for all interfaces and sensor arrangements. This ultrasonic approach sensor is relatively inexpensive, simple to use and appears to be relatively easily integrated into a VB growth furnace. The sensor is likely to be non-invasive to the crystal growth process and promises to provide significant information about the interfacial geometry and growth characteristics of difficult to grow compounds such as CdZnTe.

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