Ultrasonic transmission at solid-liquid interfaces

Haydn N. G. Wadley, Douglas T. Queheillalt and Yichi Lu

Intelligent Processing of Materials Laboratory, School of Engineering and Applied Science
University of Virginia, Charlottesville, VA 22903

Southwest Research Institute, 6220 Culebra Road, San Antonio, TX 78228-0510

ABSTRACT

New non-invasive solid-liquid interface sensing technologies are a key element in the development of improved Bridgman growth techniques for synthesizing single crystal semiconductor materials. Laser generated and optically detected ultrasonic techniques have the potential to satisfy this need. Using an anisotropic 3-D ray tracing methodology combined with elastic constant data measured near the melting point, ultrasonic propagation in cylindrical single crystal bodies containing either a convex, flat, or concave solid-liquid interface has been simulated. Ray paths, wavefronts and the time-of-flight (TOF) of rays that travel from a source to an arbitrarily positioned receiver have all been calculated. Experimentally measured TOF data have been collected using laser generated, optically detected ultrasound on model systems with independently known interface shapes. Both numerically simulated and experimental data have shown that the solidification (interfacial) region can be easily identified from transmission TOF measurements because the velocity of the liquid is much smaller than that of the solid. Since convex and concave solid-liquid interfaces result in distinctly different TOF data profiles, the interface shape (convex or concave) can also be readily determined from the TOF data. When TOF data collected in the diametral plane is used in conjunction with a nonlinear least squares algorithm, the interface geometry (i.e. position and shape) has been successfully reconstructed and ultrasonic velocities of both the solid and liquid obtained with reconstruction errors less than 5%.

Key words: Laser ultrasonic sensing, ultrasonic time-of-flight (TOF), Bridgman crystal growth, ultrasonic reconstruction.

2. INTRODUCTION

Many of today's single crystal semiconductors are grown by variants of a vertical Bridgman technique in which a cylindrical ampoule of a molten semiconductor is translated through a thermal gradient, resulting in directional solidification and the growth of a single crystal, Fig. 1.1 It has long been recognized that during crystal growth the velocity and the shape 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. the loss of single crystallinity), solute (dopant) segregation, dislocation generation, etc. They thus determine the crystals' resulting quality.2-4 The solidification rate and the solid-liquid interface shape are also sensitive functions of the internal temperature gradient (both axial and radial) during solidification.5,6 This in turn 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 and surface tension driven convection, and radiation) within the
ampoule. The solid-liquid interface’s instantaneous position, velocity and shape during crystal growth are therefore difficult to predict and uncontrolled during growth, especially for those semiconductor materials with low thermal conductivity (e.g. CdTe).\textsuperscript{4-6} The development of technologies to non-invasively sense the interface position and shape throughout the vertical Bridgman crystal growth process has therefore become a key step toward developing a better understanding of the growth process, for enabling sensor-based process control, and thus ultimately for improving the yield and quality of difficult to grow semiconductor materials like CdTe.

Ultrasonic techniques are of potential interest for this sensing need because of the significant (15 - 55\%) difference in ultrasonic velocity between the solid and liquid phases of many semiconductor materials.\textsuperscript{7-9} Thus, significant time delays and refraction occurs when ultrasound is propagated toward semiconductor solid-liquid interfaces. For single crystal solid-liquid bodies ultrasonic propagation is controlled by the magnitude and direction of the group velocity in the anisotropic solid, the solid:liquid velocity ratio, and the curvature of the interface.

Figure 1. A laser ultrasonic solid-liquid interface sensing concept for Bridgman growth of single crystal semiconductor materials.
Here, we present detailed investigations of wave propagation in single crystal solid-liquid bodies. We first study 3-D ray paths and time-of-flight (TOF) of ultrasonic rays propagating in anisotropic solid-liquid bodies. We show that ray paths are most severely bent on the crystal planes with the greatest group velocity and largest interface curvature; the sometimes severe nature of the ray bending implies a need for a precise knowledge of ray paths in order to use TOF data for interface reconstruction. It is found that ultrasonic sensing in the diametral plane is the preferred sensing configuration for interface reconstruction, since in this case the plane in which the reflected and transmitted rays propagate is known beforehand, and more ray paths are available in the plane for interface reconstruction. Furthermore, only a small set of TOF projection data is needed for determining both the interface position and its convexity. A robust nonlinear least squares algorithm in conjunction with measured TOF projection data is shown to accurately reconstruct the position/curvature of the interface and values of the velocity fields.

3. RAY PATH ANALYSIS

Consider a crystal cylinder of radius $R$, Fig. 2. The axis of the cylinder is chosen to coincide to the $x_3$ axis. Following Shah\(^1\) the outer boundary of the cylinder is expressed as:

$$\phi_c = x^T A_c x - c_c = 0$$  \hspace{1cm} (1)

where $x = (x_1, x_2, x_3)$,

$$A_c = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix} \hspace{1cm} c_c = R^2$$  \hspace{1cm} (2)

![Diagram of 3-D ray tracing in a single crystal solid-liquid body.](image)

Figure 2. An illustration of 3-D ray tracing in a single crystal solid-liquid body.
We assume the solid-liquid interface is spherical with a radius, $R_c$, centered on the $x_3$ axis. The interface, $\phi_s$, can be expressed as:

\[
\phi_s = x^T A_s x + B_s^T x + c_s = 0
\]  
(3)

where

\[
A_s = \begin{bmatrix}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 1
\end{bmatrix}
\]  
(4)

For convex interfaces,

\[
B_s = \begin{bmatrix}
0 \\
0 \\
2(R_c - h)
\end{bmatrix} \quad c_s = h^2 - 2R_c h \quad h > 0
\]  
(5)

For concave interfaces

\[
B_s = \begin{bmatrix}
0 \\
0 \\
-2(R_c + h)
\end{bmatrix} \quad c_s = h^2 + 2R_c h \quad h < 0
\]  
(6)

In (5) and (6), $h$ defines the interface convexity. The interface is flat when $h = 0$ ($c_s = 0$). Using $R$ and $h$, the interface radius $R_c$ can be expressed as:

\[
R_c = \frac{R^2 + h^2}{2h}
\]  
(7)

Let $S$ be a prescribed source point and $l$ the initial ray direction vector from the source point, $S$. $P_1$ and $P_2$ are the first and the second intersection points of the ray path with the interface surface. $R$ is the receiver point, while $t_1$ and $r_1$ are the transmitted and reflected ray direction vectors at $P_1$, $t_2$ is the transmitted ray direction vector at $P_2$. Thus,

\[
P_1 = S + L_1 l
\]  
(8)

where $L_1$ is the distance from $S$ to $P_1$.

Since $P_1$ is on the interface it satisfies Eq. (3):

\[
P_1^T A_s P_1 + B_s^T P_1 + c_s = 0
\]  
(9)

Substituting Eq. (8) into Eq. (9) yields

\[
L_1^2 (U^T A_s l) + L_1 (2S^T A_s l + B_s^T l) + (S^T A_s S + B_s^T S + c_s) = 0
\]  
(10)

Equation (10) yields two roots for $L_1$. When both roots are complex the ray path does not intersect the interface; when both roots are real and positive, the smaller root is the correct solution for $L_1$; when one
root is positive and another is negative, the positive root is the solution for $L_1$ (the negative root is for the ray travelling in the opposite direction with respect to $I$); when both roots are real and negative, then the ray travels away from the interface (it does not intersect the interface).

The normal $N_1 = (n_1, n_2, n_3)$ at $P_1$ to the interface can be obtained using the gradient of the interface:

$$N_1 = \frac{\nabla \phi_s}{\sqrt{(\nabla \phi_s)^T(\nabla \phi_s)}}$$

(11)

where

$$\nabla \phi_s = 2A_s P_1 + B_s$$

(12)

is the interface gradient at $P_1$. $N_1$ is chosen such that it always points into the liquid ($n_3 > 0$).

The incident angle $\alpha_{in1}$ at $P_1$ to the interface can then be expressed as:

$$\cos \alpha_{in1} = -I \cdot N_1$$

(13)

To determine the reflected and transmitted ray paths, we notice that when a ray is incident upon an interface, both the reflected and transmitted rays propagate in the plane defined by the incident vector, $I$, and the normal to the interface, $N_1$, at the intersection point $P_1$. If $r$ is an arbitrary vector in this plane then this plane is given as:

$$(r - P_1) \cdot (I \times N_1) = 0$$

(14)

where $(I \times N_1)$ is the normal to the plane at $P_1$.

To determine the reflected and transmitted ray paths at $P_1$, we need to determine the expression of the stiffness tensor in the plane defined by Eq. (14). We introduce a local coordinate system $(x'_1, x'_2, x'_3)$ in which the $x'_2$ axis is in the $N_1$ direction, the $x'_3$ axis is in the $(I \times N_1)$ direction, and the $x'_1$ axis is in the direction defined by $[N_1 \times (I \times N_1)]$ which is tangential to the interface. Using the directional cosines between $(x_1, x_2, x_3)$ and $(x'_1, x'_2, x'_3)$ the expression of the stiffness tensor in the local $(x'_1, x'_2, x'_3)$ coordinate system can now be obtained following the procedure described by Auld. Knowing the plane in which the incident, reflected, and transmitted rays propagate and the expression of the elastic stiffness tensor in the local $(x'_1, x'_2, x'_3)$ coordinate system, the problem of determining the reflected and transmitted ray paths becomes 2-D and can be solved using the procedures described in a previous publication.

For a convex interface and a source point above the interface, a transmitted ray path may intersect the interface again at $P_2$; the point $P_2$ is obtained in exactly the same way as $P_1$. We call this type of ray a doubly transmitted ray. Because $v_s > v_h$, the refraction angle in the liquid is smaller, and there is only one intersection point on the solid-liquid interface for a convex interface when the source point is below the interface or when the interface is concave. This type of ray is called a singly transmitted ray. Using $t_1$ and $r_1$, the second intersection point $P_2$ and the reflected ray point $R$ leaving $P_1$ can be written
\[ P_2 = P_1 + L_2 t_1, \quad R = P_1 + L_R r_1 \]

where \( L_2 \) is the distance from \( P_1 \) to \( P_2 \), \( L_R \) is the length of the reflected ray leaving from \( P_1 \).

All rays eventually intersect the cylinder at \( P_3 \). The distance between \( P_2 \) and \( P_3 \) when there is a second intersection point on the solid-liquid interface, or between \( P_1 \) and \( P_3 \) when there is only a single intersection point can be determined using Eq. (1). The reflected ray path is then given by \( S, P_1 \) and \( R \), the singly transmitted ray path by \( S, P_1, P_3 \), while the doubly transmitted ray path is given by \( S, P_1, P_2, P_3 \).

To numerically simulate wave propagation we use the material constants for Germanium (Ge) because its elastic constants as functions of temperature were available for both the solid and the liquid. The analysis can be extended to any anisotropic materials (cubic or non cubic) with know elastic properties. We use the values of elastic constants of Ge at 900°C:\(^{18}\)

\[ c_{11} = 108.45 \, \text{GPa}, \quad c_{12} = 108.45 \, \text{GPa}, \quad c_{44} = 108.45 \, \text{GPa} \]

At this temperature the density \((\rho)\) of the solid is 5.26 g/cm\(^3\).\(^{19}\) The sound velocity in the liquid (at a temperature just over Ge’s melting temperature of 937°C) is 2.71 mm/μs.\(^{20}\) In all cases \( R = 37.5 \, \text{mm} \).

For a convex interface shape, \( h = 15 \, \text{mm} \), with the source point being located at \((0, -R, 15)\), Figs. 3a and 3b show reflected and doubly transmitted ray paths. When the initial ray directions \( l \) are in the diametral plane all ray paths stay in the same plane, since the interface is symmetric with respect to the plane and the normal to the interface at the intersection is also in this plane. When the initial ray directions are not in the diametral plane the reflected and transmitted rays travel away from the diametral plane. Notice most forward propagating doubly transmitted ray paths travel in the diametral plane; the doubly transmitted rays that do not travel in the diametral plane propagate backward due to severe ray bending. Figs. 4a and 4b show singly transmitted ray paths for a convex interface shape, \( h = 15 \, \text{mm} \), with the source point being located at \((0, -R, -15 \, \text{mm})\). No doubly transmitted ray paths exist for this case. Similar results were observed for concave interface shapes, \( h < 0 \).

When the initial ray directions are in the diametral plane and the interface curvature is symmetric with respect to this plane, rays always travel in the diametral plane. Thus, the plane in which reflected and refracted rays travel is known \textit{a priori}. When the initial ray directions are not in the diametral plane, the plane in which reflected and refracted rays travel is not known beforehand and needs to be determined in order to obtain the reflected and transmitted ray paths. The orientation of this plane \textit{depends upon} the interface curvature and the incident ray direction. More rays travel in the diametral plane than in other planes that can be used for the interface sensing purpose. Thus for ultrasonic characterization of the solid-liquid interface using measured ultrasonic TOF data, positioning the ultrasonic source and receiver in the diametral plane should always be the first choice. There are no fundamental difficulties to using ray paths in non diametral planes for interface sensing, it is just more complex, time consuming, and, possibly less accurate.
Figure 3. Ray paths for a convex interface, $h = 15$ mm, $s_3 = 15$ mm. (a) reflected ray paths; (b) doubly transmitted ray paths.

Figure 4. Transmitted ray paths for a convex interface, $h = 15$ mm, $s_3 = -15$ mm. (a) the view direction is in the x axis; (b) the view direction is in the y axis.

4. PREDICTED TIME-OF-FLIGHT PROJECTS

Ultrasonic TOF projection data are affected by the crystal orientation, the liquid and the solid velocities as well as the interface curvature. To use ultrasonic TOF data for interface sensing, we are interested in ray paths that intersect the solid-liquid interface and thus carry information about the interface. We first examine TOF projection data for a convex interface, $h = 15$ mm, when the source is above the interface, $s_3 = 15$ mm. Fig. 5a shows diametral TOF projection data for reflected, singly and doubly transmitted ray paths where the receiver position is translated up and down in the diametral plane. The TOF of reflected ray paths increases with $r_3$ due to the longer propagation distance, the TOF of the doubly transmitted ray paths coincides with that of the singly transmitted ray paths at $r_3 = 0$, which is the minimum of the projection data. Fig. 5b shows circumferential TOF projection data of reflected and singly transmitted ray paths as functions of receiver angle $\alpha$ as defined in Fig. 4 of ref. [17] for several different vertical receiver
positions, $r_3$. When the source point is above the interface most forwardly propagating doubly transmitted rays travel in the diametral plane, Fig. 3b, a circumferential scan of the receiver point around the ampoule cylinder is unlikely to detect doubly transmitted signals except near the diametral plane. The circumferential TOF values of the reflected ray paths increase with increasing $r_3$ and decreasing $\theta d$ due to the longer propagation distance. The same explanation also applies to the singly transmitted rays. Due to the screen effect of the interface, only doubly transmitted rays exist near the diametral plane in the region $0 < r_3 < 15$ mm. Fig. 6a and 6b show diametral and circumferential TOF data for a convex interface where the source is below the interface at $(0, -R, -15)$. In the diametral plane the TOF of the direct rays has a maximum at $r_3 = -15$ mm where the solid velocity is minimum. The TOF of transmitted rays increases with the receiver position due to longer traveling distance. The behavior of circumferential TOF is relatively simple, the higher the receiver position, the greater the TOF value. Again, similar results were observed for concave interface shapes.

Figure 5. A convex interface, $h = 15$ mm, $s_3 = 15$ mm. (a) Diametral TOF data as a functions of the vertical receiver coordinate; (b) circumferential TOF data as functions of the receiver angle.

Figure 6. A convex interface, $h = 15$ mm, $s_3 = -15$ mm. (a) Diametral TOF data as functions of the vertical receiver coordinate; (b) circumferential TOF data as functions of the receiver angle.
An inspection of TOF projection data reveals that both the values and the shape of TOF curves are different for different sensing configurations and interface curvature. For instance, concave interfaces are characterized by a steep hump in circumferential TOF curves near $\alpha = 0^\circ$. For the convex interface when the source point is above the interface, singly transmitted rays can be detected circumferentially near the interface, while for the concave interface, the singly transmitted rays travel down into the solid cylinder except those in the diametral plane. Thus, measurable TOF data for singly transmitted rays near an interface would indicate a convex interface shape. On the diametral plane determining whether the convex interface is above or below the source point can be easily done by inspecting the TOF curves, Figs. 5a and 6a. For concave interfaces, the TOF values for rays that travel below the interface when the source point is above the interface (transmitted rays) are much larger than those when the source point is below the interface (direct rays, not shown), while the TOF values for rays that travel above the interface when the source point is above the interface (reflected rays) are much smaller that those when the source point is below the interface (transmitted rays, not shown). If we compare convex and concave interfaces where $s_3 > 0$, we find that for a convex interface the TOF of transmitted rays increases with decreasing $r_3$, while for a concave interface the TOF of transmitted rays decreases with decreasing $r_3$.

5. EXPERIMENTAL TIME-OF-FLIGHT PROJECTION DATA

To experimentally explore the use of a laser ultrasonic sensor concept for monitoring the solid-liquid interface location and shape we used a combination of ray path analysis and testing of a model (isotropic) system (where interface geometries are precisely known). 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. 17

5.1 Model System and Experimental Procedure

The bench-top model consisted of water and solid PMMA contained in a cylindrical aluminum (2024-T6) "ampoule" because opaque ampoules such as pyrolytic boron nitride (PBN) are sometimes used during crystal growth. By machining the end of the PMMA, the interfacial curvature could be varied from convex to concave (viewed toward the liquid). 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 0.5mm in diameter. Thus, the source power density was ~1500MW/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.
5.2 Experimental Time-of-Flight Projects

Measured TOF projections along the diametral plane for convex interfaces \((h = 2, 5 \text{ and } 10 \text{ mm})\) are shown in Fig. 7a and 7b for a receiver point located in the liquid \((z_r = 15\text{mm})\) and the solid \((z_r = -15\text{mm})\) respectively. For the experiments it was more conducive to fix the receiver point \((z_r)\) and scan the source point \((z_s)\). For the case where the receiver is located in the liquid, Fig. 7a, and the source also in the liquid \((z_s > 0)\) two wavefront arrivals corresponding to doubly transmitted and non refracted (direct) rays were observed. From Fig. 7a it is clear that for most sensor arrangements the TOF of doubly transmitted rays is always smaller than those of non direct rays and an energy loss was observed in the ultrasonic waveforms acquired. 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 transmitted rays. Now consider the receiver located in the solid and the source in the liquid \((z_s > 0)\), Fig. 7b, only one wavefront arrival was observed, corresponding to singly transmitted rays. Again as the source was scanned into the solid \((z_s < 0)\), only one wavefront arrival was observed, corresponding to non refracted (direct) rays. The measured TOF's are in good agreement with those predicted by the 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 transmitted and direct rays to singly transmitted rays as the source was scanned from the liquid to the solid. Also the magnitude of the TOF traces of singly and doubly transmitted rays decreased and there was an increase in the slope of doubly transmitted rays as the interface convexity \((h)\) increased. When the source was located in the solid, there was a transition of singly transmitted rays to direct rays as the source was scanned from the liquid to the solid. Again the magnitude of the TOF traces of singly transmitted rays decreased and slope increased as the interface convexity \((h)\) increased. Therefore, the convexity of convex interfaces can be qualitatively determined from direct observation of the TOF projection data, whereas the location \((z_i)\) can be quantitatively evaluated. Similar results were observed with concave interfaces.

![Figure 7. Ultrasonic time-of-flight projection data for convex interfaces, \(h = 2, 5 \text{ and } 10\text{ mm}\), for a receiver located in (a) the liquid \((z_r = 15\text{mm})\) and (b) the solid \((z_r = -15\text{mm})\).](image)
5.3 Interface Curvature Reconstructions

In a crystal growth application, the position 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.\textsuperscript{21} For crystal growth applications, approaches that can be used with sparse data that exploits the often significant \textit{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.

We assume the model geometry is of the form shown in Fig. 2, where \( h, z_p, v_l \) and \( v_s \) are all unknown. For the refracted ray path model, the TOF depends non-linearly on the interface convexity \( h \), the interface location \( (z_l) \), the liquid \( (v_l) \) and solid \( (v_s) \) velocities. The mean-square error is given by

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

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 diametral TOF projection data a Levenberg-Marquardt nonlinear least-squares reconstruction method was used.\textsuperscript{22} The nonlinear least-squares algorithm returned the best-fit (i.e. reconstructed) parameters \( (h, z_p, v_l, v_s) \) by converging upon the interface that minimized \( \chi^2 \).

The reconstructed interface location \( (z_l) \) and convexity \( (h) \) obtained using the nonlinear least-squares method for convex interfaces with the receiver located in the liquid were within \( \pm 0.95\text{mm} \) and \( \pm 0.94\text{mm} \), respectively for all interfaces \( (h = 2, 5 \text{ and } 10\text{mm}) \) and receiver locations \( (z_r = 5, 10, 15 \text{ and } 20\text{mm}) \). 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 \( (z_l) \) and convexity \( (h) \) were within \( \pm 0.93\text{mm} \) and \( \pm 1.32\text{mm} \), respectively for all interfaces \( (h = 2, 5 \text{ and } 10\text{mm}) \) and receiver locations \( (z_r = -5, -10, -15 \text{ and } -20\text{mm}) \). Again, the deduced liquid and solid velocities were within 4.9\% (for the liquid) and 2.4\% (for the solid) of the actual velocities.

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 accurately predict the interface position and shape. Using the general form of these solutions with free parameters \( (h, z_p, v_l, v_s) \) 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 position, convexity) and velocity fields from diametral 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.
6. CONCLUSIONS

Three-dimensional ray path, wavefront, and TOF simulations have been conducted on cylindrical single-crystal solid-liquid bodies. It has been demonstrated that positioning the sensor near the diametral plane is the most promising sensing configuration for solid-liquid interface sensing because: (1) the plane in which reflected and transmitted rays propagate is known, and (2) more ray paths with different properties (reflected, transmitted) are available in this plane for interface reconstruction. There is no fundamental difficulty to use ray paths in non diametral planes, but it is more complicated to use them and fewer rays are available for interface reconstruction. The TOF projection data for different sensing configurations and different interface curvature are distinctively different, indicating the potential of laser ultrasonic sensors for solid-liquid interface determination during single crystal growth.

7. ACKNOWLEDGMENTS

This work has been performed as a part of the research of the Infrared Materials Productibility Program conducted by a consortium that includes Johnson Matthey Electronics, Texas Instruments, II-VI Inc., Loral, the University of Minnesota, and the University of Virginia. We are grateful for the many helpful discussions with our colleagues in these organizations. The consortium work has been supported by ARPA/CMO under contract MD A972-91-C-0046 monitored by Raymond Balcerak.

8. REFERENCES