Three-dimensional wave propagation through single crystal solid–liquid interfaces

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Large differences in the ultrasonic velocity of the solid and liquid phases of semiconductors have stimulated an interest in the use of laser ultrasonic methods for locating and characterizing solid–liquid interfaces during single crystal growth. A previously developed two-dimensional ray tracing analysis has been generalized and used to investigate three-dimensional ultrasonic propagation across solid–liquid interfaces in cylindrical bodies where the receiver is located at an arbitrary position relative to the source. Numerical simulations of ultrasonic ray paths, wavefronts, and time of flight have indicated that ultrasonic sensing in the diametral plane is a preferred sensing configuration since the transmitted, reflected, and refracted rays all propagate in this plane, significantly simplifying analysis of the results. While other sensing configurations can also provide information about solid–liquid interfaces, they require a more complicated analysis because the planes in which reflected and refracted rays propagate are not known a priori, and fewer ray paths are accessible for interface interrogation because of large refractions. © 1998 Acoustical Society of America. [S0001-4966(98)00203-3]

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INTRODUCTION

The Bridgman growth of single crystal semiconductor materials with low thermal conductivity often results in poor crystal quality and a low yield of useful wafers for microelectronic, optoelectronic, or infrared detector applications. The crystal quality is thought to be largely controlled by the velocity and curvature of the solid–liquid interface during the growth process. The best crystal quality usually results from a planar or slightly convex interface shape that propagates at a uniform slow rate along the axis of the boule. Thus sensing and control of the solid–liquid interface during crystal growth appears to be a key step toward improving crystal quality. In most systems of interest, large decreases in ultrasonic velocity accompany melting. The large difference in the ultrasonic velocity between the solid and the liquid phases of most semiconductor materials, together with the recent emergence of noninvasive laser ultrasonics, have stimulated an interest in using ultrasonics to monitoring solid–liquid interfaces during single crystal growth.

In previous work, the severe ray bending associated with the large velocity change at liquid–solid interfaces has been shown to require a detailed ray tracing analysis to use ultrasonic time of flight (TOF) projection data for characterizing a solid–liquid interface. Using a two-dimensional analysis, a detailed study of ray paths, wavefronts, and TOF of ultrasound signals propagating on either the transverse or diametral planes of liquid single crystal solid bodies was reported. Numerical simulations indicated that the magnitude and direction of the group velocity, the solid–liquid velocity ratio, and the curvature of the interface together controlled the ray bending behavior, and thus determined ultrasonic paths across the interface. These theoretical studies indicated that when the ray paths through an interface could be computed, the interface location and curvature could be reconstructed from sparse ultrasonic TOF data collected on the diametral plane. Laser ultrasonic experiments conducted on benchtop models have recently confirmed that the interface location and curvature can indeed be reconstructed from the measured TOF data using a simple nonlinear least squares algorithm, an axisymmetric interface model, and the ray tracing algorithm, developed in Ref. 10.

The sensor configurations explored to date have been constrained by our limited understanding of wave propagation at anisotropic solid–liquid interfaces. The availability of only 2-D wave propagation algorithms required the placement of sources and receivers on the transverse or diametral plane where both the transmitted and refracted rays must propagate. Here, we generalize the 2-D wave propagation analysis to 3-D, and allow the receiver to be located at arbitrary positions relative to the source. Similar to our previous work, we study only the (fastest) quasilongitudinal wave mode. The diffracted rays have been studied in our previous work. We employ a matrix/vector approach for the 3-D ray tracing analysis that has previously been developed for isotropic materials. The key step in analyzing the 3-D wave propagation in anisotropic materials is shown to be the determination of the usually unknown plane in which both the reflected and transmitted rays propagate. With this plane determined, the procedure for constructing elastic stiffness tensors and the Christoffel equations that govern wave propagation velocity in the local coordinate systems of the interface then become the same as the 2-D case. Numerical simulations of ray paths, wavefronts, and TOF are presented for both convex and concave interface shapes with the ultrasonic source/receiver located either above or below the interface. It
will be shown that rays traveling in nondiametral planes can be used for interface sensing. However, because the plane in which the reflected and transmitted rays propagate is not known beforehand (and needs to be determined at each intersection point of the ray path with the interface), analysis of TOF data is considerably more complex. When diametral plane access is available, it appears to be the preferred approach for sensing axisymmetric interfaces.

I. RAY PATH ANALYSIS

Consider a circular crystal cylinder of radius $R$ (Fig. 1). The axis of the cylinder is chosen to coincide with the $x_3$ axis of our coordinate system. Following Shah\textsuperscript{12} the outer boundary surface of the cylinder, $\phi_c$, can be expressed as:

$$\phi_c = x^T A_c x - c_c = 0,$$

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

$$A_c = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad c_c = R^2.$$  

(2)

We assume that the solid–liquid interface is a spherical cap with a radius, $R_c$, centered on the $x_3$ axis. The intersection of the solid–liquid interface and the cylinder lies in the $x_1-x_2$ plane. Thus the interface is symmetric with respect to both the $x_1-x_3$ and the $x_2-x_3$ planes. The interface, $\phi_s$, can be expressed as:

$$\phi_s = x^T A_s x + B_s^T x + c_s = 0,$$

where

$$A_s = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad B_s = \begin{bmatrix} 0 \\ 0 \\ 2(R_c - h) \end{bmatrix}, \quad c_s = h^2 - 2R_c h, \quad h > 0.$$  

(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 Eqs. (5) and (6), $h$ defines the interface convexity (Fig. 1). The interface is flat when $h = 0 (c_s = 0)$. In terms of the cylinder radius, $R$, and the convex interface convexity, $h$, the interface radius of curvature $R_c$, can be expressed as:

$$R_c = \frac{R^2 + h^2}{2h}.$$  

(7)

We let $S$ be a prescribed source point, and $I$ be the initial ray direction vector pointing from the source point. $S$, $P_1$ and $P_2$ are then the first and the second intersection points of a general ray path propagating through the interface to a receiver point $R$; $t_1$ and $r_1$ are then the transmitted and reflected ray direction vectors at $P_1$, while $t_2$ is the transmitted ray direction vector at $P_2$. Thus

$$P_1 = S + L_1 I,$$

(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

$$L_1^2 (I^T A_s I) + L_1 (2S^T A_s I + B_s^T I) + (S^T A_s S + B_s^T S + c_s) = 0.$$  

(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 one is the correct solution for $L_1$; when one root is positive and the other is negative, the positive one is the solution for $L_1$ (the negative root is for the ray traveling 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 from 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 gradient of the interface at $P_1$. The normal of the interface, $N_1$, is chosen such that it always points into the liquid ($n_3 \geq 0$).

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

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

(13)

To determine the plane in which reflected and transmitted rays propagate, 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, \( \mathbf{l} \), and the normal to the interface, \( \mathbf{N}_1 \), at the intersection point \( \mathbf{P}_1 \).\(^{13-19} \) If \( \mathbf{r} \) is an arbitrary vector in the plane, this plane is given as:

\[
(\mathbf{r} - \mathbf{P}_1) \cdot (\mathbf{l} \times \mathbf{N}_1) = 0,
\]

where \( (\mathbf{l} \times \mathbf{N}_1) \) is the normal to the plane at \( \mathbf{P}_1 \).

To determine the reflected and transmitted ray paths emitting from \( \mathbf{P}_1 \) and propagating in the plane defined by Eq. (14), we first need to determine an expression for the stiffness tensor in the plane. We introduce a local coordinate system \((x_1', x_2', x_3')\) in which the \( x_3' \) axis is in the \( \mathbf{N}_1 \) direction, the \( x_1' \) axis is in the \((\mathbf{l} \times \mathbf{N}_1)\) direction, and the \( x_2' \) axis is in the direction defined by \( [\mathbf{N}_1 \times (\mathbf{l} \times \mathbf{N}_1)] \) 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.\(^{10,14} \)

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 thus be solved using the procedures described in a previous publication.\(^{10} \)

When the source point is in the liquid, the direction vectors of the transmitted rays in the solid, \( \mathbf{t}_1 \), and reflected rays in the liquid, \( \mathbf{r}_1 \), can be shown to be

\[
\mathbf{t}_1 = \frac{v_g}{v_l} \left[ \frac{v_g}{v_l} \cos(\alpha_{in1}) - \cos(\alpha_{tr1}) \right] \mathbf{N}_1,
\]

\[
\mathbf{r}_1 = 1 + 2 \cos(\alpha_{in1}) \mathbf{N}_1,
\]

where \( v_g \) is the magnitude of the group velocity in the solid, \( \alpha_{in1} \) is the refraction angle in the solid, Fig. 1. When the source point is in the solid, all rays intersecting the interface are transmitted into the liquid due to the smaller liquid velocity and the smaller refraction angle. The refraction direction vector in the liquid in this case is:

\[
\mathbf{t}_1 = \frac{v_g}{v_l} \left[ \frac{v_g}{v_l} \cos(\alpha_{in1}) - \cos(\alpha_{tr1}) \right] \mathbf{N}_1.
\]

For a convex interface and a source point above the interface, a transmitted ray path may intersect the interface again at \( \mathbf{P}_2 \); the point \( \mathbf{P}_2 \) is obtained in exactly the same way as \( \mathbf{P}_1 \). This type of ray is called a doubly transmitted ray. When the source point is below a concave interface, there is only one intersection point on the solid–liquid interface due to the interface curvature and the smaller refraction angle in the liquid resulting from the smaller liquid velocity. The ray with a single intersection point on the interface is called singly transmitted ray. Using \( \mathbf{t}_1 \) and \( \mathbf{r}_1 \), the second intersection point \( \mathbf{P}_2 \) and the reflected ray point \( \mathbf{R} \) leaving \( \mathbf{P}_1 \) can be written as

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**FIG. 2.** Ray paths for a convex interface, \( h = 15 \text{ mm}, x_3 = 15 \text{ mm} \). (a) Reflected ray paths; (b) singly transmitted ray paths; (c) doubly transmitted ray paths.

**FIG. 3.** Transmitted ray paths for a convex interface, \( h = 15 \text{ mm}, x_3 = -15 \text{ mm} \). (a) The viewing direction is parallel to the \( x_1 \) axis; (b) the viewing direction is parallel to the \( x_2 \) axis.
II. NUMERICAL IMPLEMENTATION

To numerically simulate wave propagation we use the material constants of germanium (Ge) because its elastic constants as functions of temperature are available for both the solid and the liquid. The analysis can easily be extended to any anisotropic material (cubic or noncubic) with known elastic properties. We use the values of elastic stiffness constants, $c_{ij}$, of Ge at 900 °C (Ref. 20) to represent the solid.

$$c_{11} = 108.45 \text{ GPa}, \quad c_{12} = 108.45 \text{ GPa},$$

$$c_{44} = 108.45 \text{ GPa}.$$

At this temperature the solid density $\rho = 5.26 \text{ g/cm}^3$ (Ref. 21). The sound velocity in the liquid (at a temperature just above the Ge melting temperature of 937 °C) is taken to be 2.71 mm/μs. In all cases $R = 37.5$ mm.

To analyze a representative convex interface shape, we take $h = 15$ mm. For a source point above the interface at $(0, -R, 15)$ mm, Fig. 2(a), (b), and (c) shows reflected, singly transmitted, and doubly transmitted ray paths. When the initial ray directions, I, are in the diametral plane, all ray paths remain in the plane. When the initial ray directions are not in the diametral plane the reflected and transmitted rays travel away from the diametral plane. This arises because when the source point is in the liquid, the convex liquid–solid interface acts as a divergent lens to ray paths (see Fig. 6 of Ref. 10 for rays traveling in the 2-D transverse plane of a liquid cylinder containing a solid core). Notice that most forward propagating doubly transmitted ray paths are in the diametral plane with a narrow incident angle region; the doubly transmitted rays that do not propagate in the diametral plane propagate backward due to severe ray bending.

The singly transmitted ray paths for the same convex interface but with a source located below the interface at $(0, -R, -15)$ are shown in Fig. 3(a) and (b). For this configuration, all rays that intersect the interface are refracted into the liquid due to the smaller liquid velocity. No doubly
transmitted ray paths exist for this configuration. Rays are seen to again travel in the diametral plane when the initial ray direction vector is also in the diametral plane. When the initial ray directions are not in the diametral plane, the ray paths are bent by the interface toward the diametral plane, similar to rays traveling in the transverse plane of a solid cylinder containing a liquid core (see Fig. 5 of Ref. 10).

Figure 4(a) and (b) shows reflected and singly transmitted ray paths, for a concave interface with \( h = 15 \text{ mm} \) and a source point above the interface at \((0, -R, 15 \text{ mm})\). The reflected ray paths travel toward the diametral plane due to the interface curvature, while the transmitted ray paths are bent away from the diametral plane due to the larger refraction angle in the solid. When the source point is below the interface at \((0, -R, -15 \text{ mm})\) [Fig. 5(a) and (b)] the transmitted ray paths are bent toward the diametral plane due to the smaller refraction angle in the liquid. There are no doubly transmitted rays since all rays that intersect the interface are refracted into the liquid due to the smaller refraction angle in the liquid.

With the ray paths fully analyzed, we can draw several important conclusions concerning optimal laser ultrasonic sensing configurations. When the initial ray directions are in the diametral plane and the interface curvature is symmetric with respect to the plane, rays always travel in the diametral plane. Thus the plane in which reflected and transmitted rays travel is known \textit{a priori} and it is straightforward to then determine the ray path between prescribed source and receiver points. When the initial ray directions are not in the diametral plane, the planes in which reflected and transmitted rays travel are not known beforehand and must be determined in order to obtain the reflected and transmitted ray paths. The orientation of the plane depends on the interface curvature and the incident ray direction vector and is not
known a priori. The bending of rays occurs near the solid-liquid interface and, for interface reconstruction purposes, this is easier to account for in the diametral plane than in other planes. Thus for ultrasonic characterization of a solid-liquid interface, positioning the ultrasonic source and receiver in the diametral plane should always be the preferable sensor approach. There are no fundamental difficulties to using ray paths in nondiametral planes for interface sensing; it is just a more complex and time consuming task. In fact, the very large refractions in nondiametral plane configurations imply a high sensitivity to the exact interface geometry, and so this off-diametral approach might be pursued if additional curvature reconstruction accuracy were needed.

III. WAVEFRONT SURFACES

Ultrasonic signals can be detected only at points where the wavefronts intersect the crystal surface. Wavefronts are obtained by connecting points along ray paths with the same traveling time. Here we only show wavefronts for a convex interface shape, \( h = 15 \) mm, where the source point is above the interface at \((0, -R, 15)\). Figure 6(a) shows the wavefronts for the direct rays in the liquid. The wavefronts are spherical, centered at the source point since the liquid velocity is isotropic. Notice that the bottom edge of the wavefront after \(20\mu s\) of propagation does not touch the interface due to the screening effect of the interface. Figure 6(b) shows wavefronts for the reflected rays that are reflected by the solid-liquid interface into the liquid region. Notice that the reflected wavefront at \(10\mu s\) intersects the ampoule cylinder at only two small separate sections. The wavefront for singly transmitted rays after \(10\mu s\) propagation is shown in Fig. 6(c). It has a ridge shaped area below the source, which is due to the smaller group velocity on the \((100)\) crystal plane of this cubic material. Due to the high velocity in the solid, the singly transmitted wavefront at \(20\mu s\) has propagated out of the region shown in Fig. 6(c). The doubly transmitted wavefronts are shown in Fig. 6(d). These wavefronts have larger \(y\) values than the corresponding wavefronts for the reflected rays because the doubly transmitted rays have traveled a significant distance in the solid which has a higher velocity than the liquid. The relatively thick middle portion and the thin wings of the wavefronts in Fig. 6(d) indicate that the region where it is possible to detect the doubly transmitted rays is greater near the diametral plane.

IV. TIME OF FLIGHT PROJECTIONS

To use ultrasonic TOF data for interface characterization, we are interested in ray paths that intersect the solid-liquid interface and thus carry information about the interface. To illustrate, we study the four following situations.

A. A convex interface, \(h=15\) mm, and source point above interface at \((0, -R, 15)\)

Figure 7(a) and (b) shows the diametral and transverse plane TOF projection results, respectively. In Fig. 7(b) the receiver angle, \(\alpha\), corresponds to the angle of deviation from the diametral plane projected onto the transverse plane (see Fig. 4 of Ref. 10 for additional details). The reflected, singly transmitted, and doubly transmitted signals can all be detected at the diametral point [Fig. 7(a)]. The TOF data for the reflected ray paths increase with the vertical receiver position due to the longer propagation distance and the smaller liquid velocity. TOF values of both the singly and doubly transmitted ray paths increase with the vertical distance between the source and the receiver. The minimum TOF value for the transmitted ray paths is reached when the receiver point meets the interface at \(r_3 = 0\).

Most doubly transmitted rays travel in the diametral plane [Fig. 2(c)] therefore a circumferential scan of the receiver around the crystal cylinder is unlikely to detect doubly transmitted signals except near the diametral plane. In the transverse planes reflected and singly transmitted TOF data increase with reducing \(|\alpha|\) because of the longer propagation distance. TOF values for the reflected ray paths increase as the receiver point is raised. Those for the singly transmitted rays decrease as the receiver point is moved closer to the interface.

B. A convex interface, \(h=15\) mm, and source point below interface at \((0, -R, -15)\)

Diametral and transverse plane TOF projection data are shown in Fig. 8(a) and (b). These are no doubly transmitted

![Wavefronts and TOF projections](image-url)
ray paths as discussed above. In the diametral plane when the receiver is below the interface, only the TOF for the direct ray paths can be detected, and its behavior is fully described by the group velocity on this $\{100\}$ plane. The TOF values for the transmitted ray paths increase with the vertical receiver position due to the longer propagation distances and a smaller liquid velocity. By comparing the diametral TOF data between Fig. 7 and Fig. 8, we see that the location of the convex interface with respect to the source point can be readily determined by translating the receiver point in the diametral plane. The transmitted transverse plane TOF data are similar to those of Fig. 7 when the source is above the interface but the TOF values are smaller.

C. A concave interface, $h=-15$ mm, and source point above the interface at $(0,-R,15)$

Diametral and transverse plane TOF projection data are shown in Fig. 9(a) and (b). The diametral reflected rays can be detected only when $r_3>s_3$ due to the interface curvature [Fig. 4(a)] while the transmitted rays can be detected only when $r_3<-15$ mm due to the large refraction angle in the solid [Fig. 4(b)]. However, except for the rays in the diametral plane, the transmitted rays intersect the outer sample surface well below the interface (due to the interfaces curvature), and so only the circumferential TOF data for reflected rays are shown in Fig. 9(b). They have a steep hump near $\alpha=0$. It is clear by comparing Fig. 7 with Fig. 9 that the interface convexity, or concave can be easily determined either from the diametral or circumferential TOF data.

D. A concave interface, $h=-15$ mm, and source point below interface at $(0,-R,-15)$

Diametral and transverse plane TOF projection data are shown in Fig. 10(a) and (b). The diametral plane TOF for the direct ray paths can be detected only when $r_3>s_3$, due again to the screening effect of the interface. The diametral TOF of transmitted ray paths can be detected only when $r_3>36$ mm because of the smaller refracted angle in the liquid [Fig. 5(a)]. By comparing the diametral TOF data in Figs. 8(a) and 10(a), we see again that the interface convexity can be determined from the diametral TOF data alone due to the different functional behavior of the projection data sets at $\alpha=0$.

In Fig. 10(b) the maximum in the TOF data for the direct ray paths is caused by the anisotropy of the solid velocity and the change in ray propagation distance as rays are swept from the $-x_1$ to the $x_2$ direction (Fig. 1). The small differences in the $r_3=-20$ and $-30$ mm data are also a consequence of the solid velocity anisotropy. By comparing Fig. 10(b) with Fig. 8(b), it can again be seen that the interface convexity can be determined from transverse plane projections when the source point is below the interface.
functions of the receiver angle $\alpha$. An inspection of TOF data in Figs. 7, 8, 9, and 10 also indicates that a greater diversity of ray paths (i.e., reflected, singly/doubly transmitted) can be detected near the interface in the diametral plane than in other nondiametral planes, indicating again that ultrasonic sensing in the diametral plane is likely to be the preferred sensing configuration.

V. CONCLUSIONS

A method has been developed for the calculation of three-dimensional ray paths, wavefront shapes, and TOF values during ultrasonic propagation through cylindrical single crystal solid–liquid bodies. It has been demonstrated that both the interface location and convexity strongly influence ultrasonic TOF projection data. Positioning sensors near the diametral plane is the preferred sensing configuration for solid–liquid interface sensing because: (1) the plane in which reflected and transmitted rays propagate is known beforehand; and (2) more ray paths (reflected, transmitted, etc.) are available in this plane for interface reconstruction purposes. There is no fundamental difficulty in using ray paths in other nondiametral planes; it is just more complicated and fewer ray paths are available for interface reconstruction. The TOF projection data and different interface curvatures are distinctively different for different interface curvatures, providing a promising real time direct method for solid–liquid interface characterization during the vertical Bridgman single crystal growth.

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